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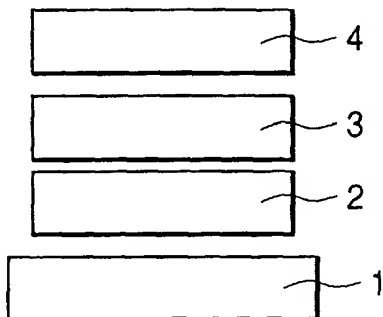
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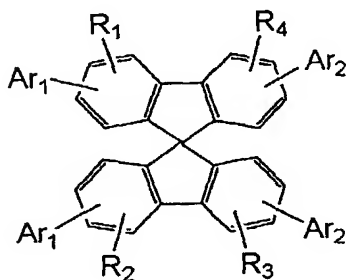
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[Continued on next page]

(54) Title: SPIRO COMPOUND AND ORGANIC LUMINESCENCE DEVICE USING THE SAME



(57) Abstract: Provided are a novel spiro compound, and an organic luminescence device using the spiro compound and having an optical output with an extremely high efficiency and a high luminance, and an extremely high durability. The spiro compound is represented by the following general formula [I]: (wherein R¹#191, R²#191, R³#191, and R⁴#191 represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a substituted amino group, a cyano group, or a halogen atom, and R¹#191, R²#191, R³#191, and R⁴#191 may be identical or different from each other; and Ar¹#191 and Ar²#191 represent a substituted or unsubstituted condensed polycyclic aromatic group or a substituted or unsubstituted condensed polycyclic heterocyclic group, which may be identical or different from each other.)



[I]



European patent (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HU, IE, IT, LU, MC, NL, PT, RO, SE, SI, SK, TR), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

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DESCRIPTION

SPIRO COMPOUND AND ORGANIC LUMINESCENCE DEVICE USING
THE SAME

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TECHNICAL FIELD

The present invention relates to a novel organic compound and an organic luminescence device using the same.

10

BACKGROUND ART

An organic luminescence device is a device where a thin film including a fluorescent organic compound or a phosphorescent organic compound is sandwiched between an anode and a cathode, an electron and a hole are injected from the respective electrodes to generate an exciton of the fluorescent compound or the phosphorescent compound, and light which is emitted when the exciton returns to the ground state is utilized.

20

According to the study of Kodak company in 1987 (Appl. Phys. Lett. 51, 913 (1987)), there has been reported a luminescence with approximately 1000 cd/m² at an applied voltage of approximately 10 V in a device having a separated-function type two-layer configuration using ITO as an anode, a magnesium-silver alloy as a cathode, an aluminum quinolinol

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complex as an electron-transporting material and a luminescent material, and a triphenyl amine derivative as a hole-transporting material. The related patents include U.S. Patent Nos. 4,539,507,
5 4,720,432, 4,885,211, and so on.

In addition, it is possible to generate luminescence in the range of ultraviolet to infrared lights by changing the type of the fluorescent organic compound, and in recent years various types
10 of compounds have been studied actively. For instance, it is described in U.S. Patent Nos. 5,151,629, 5,409,783, 5,382,477, Japanese Patent Application Laid-Open Nos. 2-247278, 3-255190, 5-202356, 9-202878, 9-227576, and so on.

15 In recent years, many studies have been conducted using phosphorescent compounds as luminescent materials and using energies in triplet excitation states. A high luminescence efficiency shown by an organic luminescence device using an
20 iridium complex as a luminescent material has been reported by a group of the Princeton University (Nature 395, 151 (1998)).

Furthermore, in addition to the organic luminescence device using the low molecular weight
25 material as mentioned above, an organic luminescence device using a conjugate polymer has been reported by a group of the Cambridge University (Nature, 347, 539

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(1990)). In this report, luminescence from a single layer is confirmed by the film formation of polyphenylene vinylene (PPV) using a coating system.

The related patents of the organic luminescence device using the conjugate polymer include U.S. Patent Nos. 5,247,190, 5,514,878, 5,672,678, Japanese Patent Application Laid-Open Nos. 4-145192, 5-247460, and so on.

In this way, the recent progress in the organic luminescence device is remarkable, and the characteristics thereof suggest the possibility of applications for various purposes, which enable the luminescence device with a high luminance, a high-speed response, and a thin and lightweight form.

However, an optical output of higher luminance or higher conversion efficiency is required under the present conditions. In addition, many problems still remain to be solved regarding the durability with respect to a change with time due to a long-term usage, deterioration caused by an atmospheric gas including oxygen, moisture, or the like, and so on. Besides, it is not still insufficient for solving problems related to the needs for luminescences of blue, green, and red having good color purity in the case of considering the applications to a full color display and so on.

On the other hand, a spiro compound having a

specific steric configuration has been attracting attention as an organic functional material in terms of the specific physical properties of the material (J. Am. Chem. Soc., vol. 110, page 5687, 1988). As
5 an example of using a spiro compound as an organic luminescence device, Japanese Patent Application Laid-Open No. 7-278537 or the like can be given, but the characteristics thereof in being used as a luminescent material or an electron-transporting
10 material are not sufficient.

DISCLOSURE OF THE INVENTION

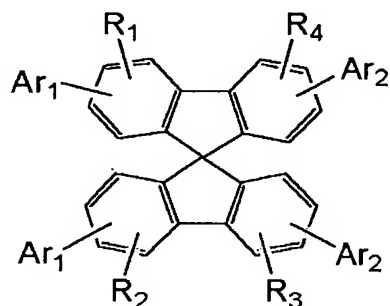
An object of the present invention is to provide a novel spiro compound.

15 Also, another object of the present invention is to provide an organic luminescence device using a specific spiro compound and having an optical output with an extremely high efficiency and a high luminance.

20 In addition, another object of the present invention is to provide an organic luminescence device having an extremely high durability. Furthermore, another object of the present invention is to provide an organic luminescence device which
25 can be easily and comparatively inexpensively produced.

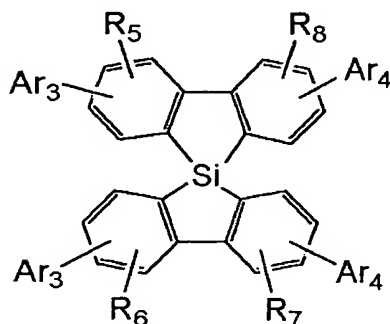
Therefore, a spiro compound according to the

present invention is represented by one of the following general formula [I] and [II]:



[I]

(where R₁, R₂, R₃, and R₄ represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a substituted amino group, a cyano group, or a halogen atom, and R₁, R₂, R₃, and R₄ may be identical or different from each other; and Ar₁ and Ar₂ represent a substituted or unsubstituted condensed polycyclic aromatic group or a substituted or unsubstituted condensed polycyclic heterocyclic group, which may be identical or different from each other); and



[I I]

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(where R_5 , R_6 , R_7 , and R_8 represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a
5 substituted or unsubstituted heterocyclic group, a substituted amino group, a cyano group, or a halogen atom, and R_5 , R_6 , R_7 , and R_8 may be identical or different from each other; and Ar_3 and Ar_4 represent a substituted or unsubstituted condensed polycyclic
10 aromatic group or a substituted or unsubstituted condensed polycyclic heterocyclic group, which may be identical or different from each other.)

Further, an organic luminescence device according to the present invention includes at least
15 a pair of electrodes including an anode and a cathode and one or a plurality of layers containing an organic compound sandwiched between the pair of electrodes, in which at least one of the layers containing the organic compound preferably contains
20 at least one spiro compound.

BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is a cross-sectional diagram that illustrates an example of an organic luminescence
25 device in accordance with the present invention.

Fig. 2 is a cross-sectional diagram that illustrates another example of the organic

luminescence device in accordance with the present invention.

Fig. 3 is a cross-sectional diagram that illustrates another example of the organic luminescence device in accordance with the present invention.

Fig. 4 is a cross-sectional diagram that illustrates another example of the organic luminescence device in accordance with the present invention.

Fig. 5 is a cross-sectional diagram that illustrates another example of the organic luminescence device in accordance with the present invention.

Fig. 6 is a cross-sectional diagram that illustrates another example of the organic luminescence device in accordance with the present invention.

BEST MODE FOR CARRYING OUT THE INVENTION

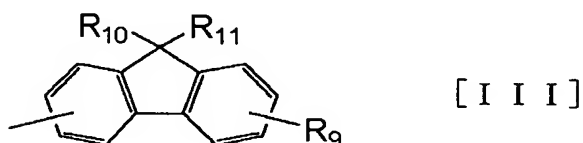
Hereinafter, the present invention will be described in detail.

At first, a spiro compound of the present invention will be described.

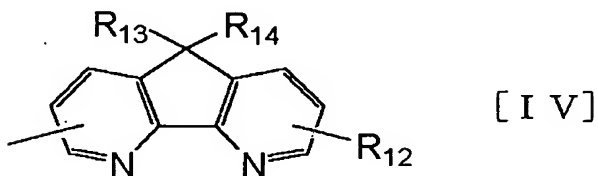
The spiro compound of the present invention is represented by the above general formula [I] or [II].

Here, at least one of Ar_1 and Ar_2 , or at least

one of Ar₃ and Ar₄ is preferably a condensed polycyclic aromatic group represented by one of the following general formula [III] to [IX]:



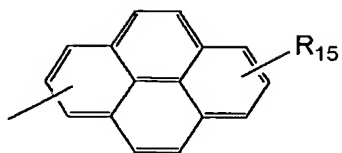
- 5 (where R₉ represents a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a substituted amino
- 10 group, a cyano group, or a halogen atom; and R₁₀ and R₁₁ represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, or a substituted or
- 15 unsubstituted heterocyclic group, which may be identical or different from each other);



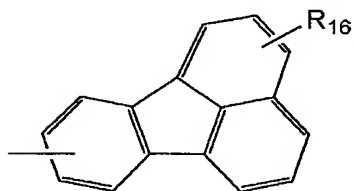
- 20 (where R₁₂ represents a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or

unsubstituted heterocyclic group, a substituted amino group, a cyano group, or a halogen atom; and R_{13} and R_{14} represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or
5 unsubstituted aralkyl group, a substituted or unsubstituted aryl group, or a substituted or unsubstituted heterocyclic group, which may be identical or different from each other); and

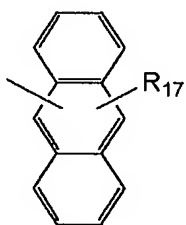
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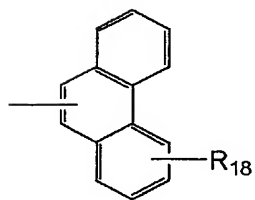
[V]



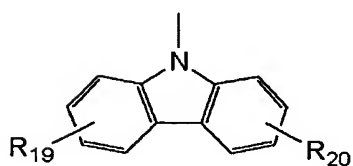
[VI]



[VII]



[VIII]



[IX]

(where R_{15} to R_{20} represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a substituted amino group, a cyano group, or a halogen

atom.)

Specific examples of substituents in the above general formula [I] to [IX] are shown below.

The alkyl group includes a methyl group, an
5 ethyl group, an n-propyl group, an iso-propyl group, an n-butyl group, a tert-butyl group, an octyl group, and the like.

The aralkyl group includes a benzyl group, a phenethyl group, and the like.

10 The aryl group includes a phenyl group, a biphenyl group, a tert-phenyl group, and the like.

The heterocyclic group includes a thienyl group, a pyrrolyl group, a pyridyl group, an oxazolyl group, an oxadiazolyl group, a thiazolyl group, a
15 thiadiazolyl group, a tert-thienyl group, and the like.

The substituted amino group includes a dimethylamino group, a diethylamino group, a dibenzylamino group, a diphenylamino group, a
20 ditolylamino group, a dianisolylamino group, and the like.

The halogen atom includes fluorine, chlorine, bromine, iodine, and the like.

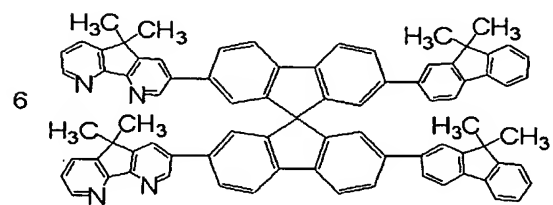
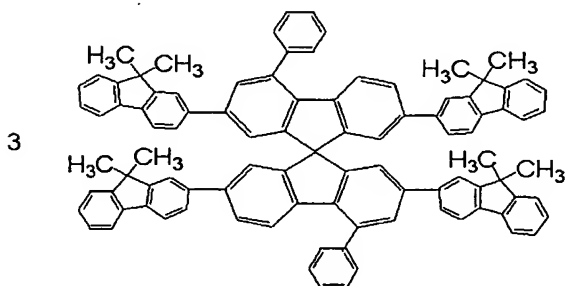
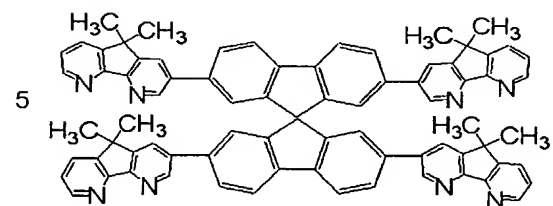
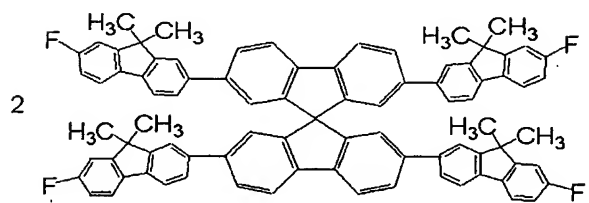
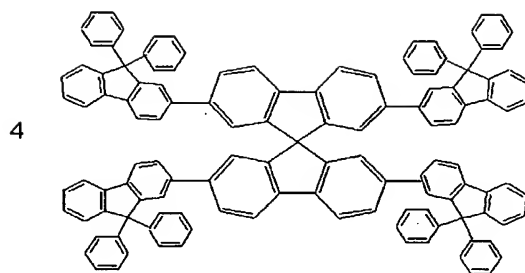
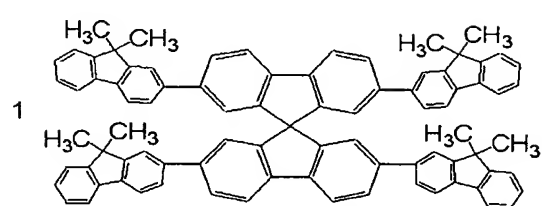
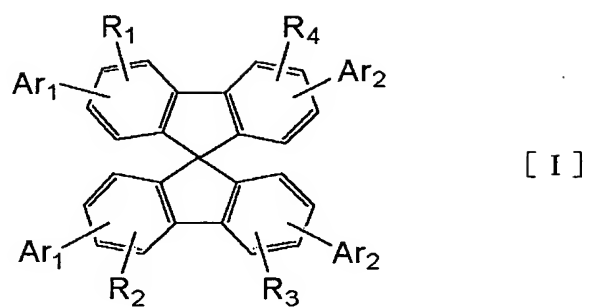
The condensed polycyclic aromatic group includes
25 a fluorenyl group, a naphthyl group, a fluoranthenyl group, an anthryl group, a phenanthryl group, a pyrenyl group, a tetracenyl group, a pentacenyl group,

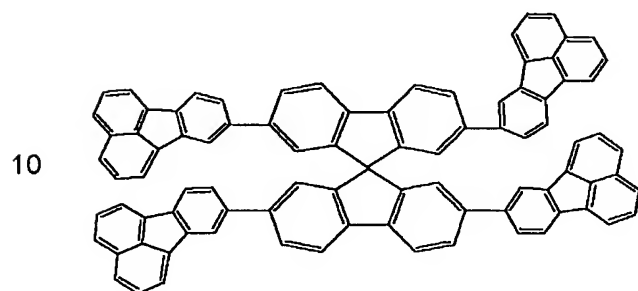
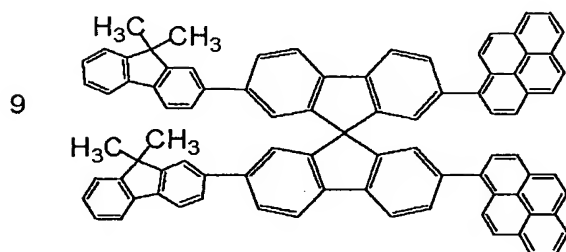
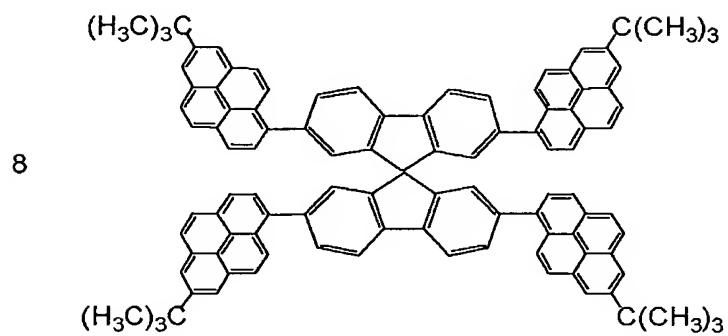
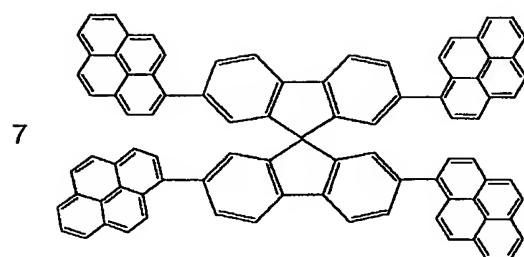
a triphenylenyl group, a perylenyl group, and the like.

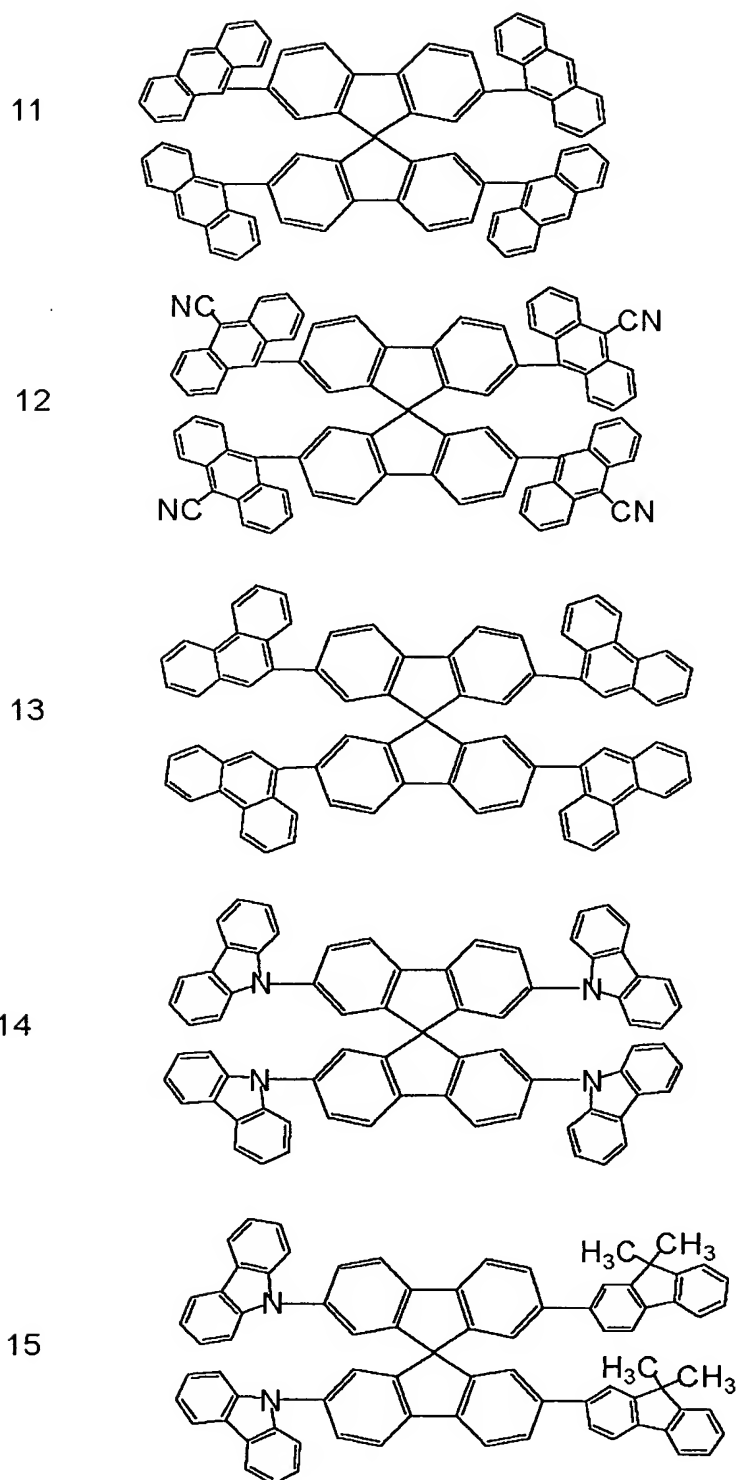
The condensed polycyclic heterocyclic group includes a carbazolyl group, a diazafluorenyl group,
5 an acridinyl group, and the like.

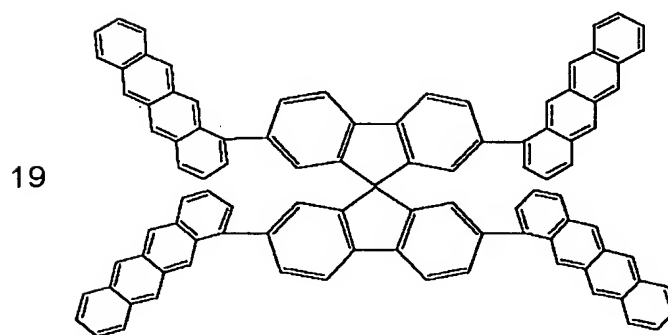
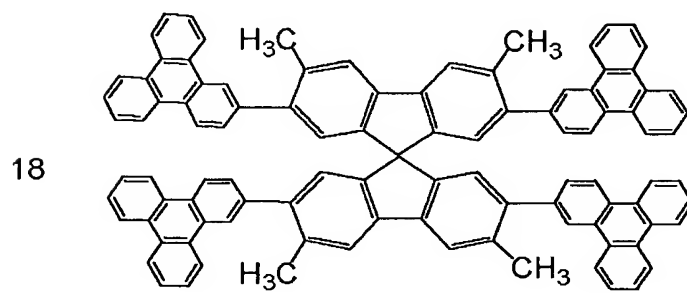
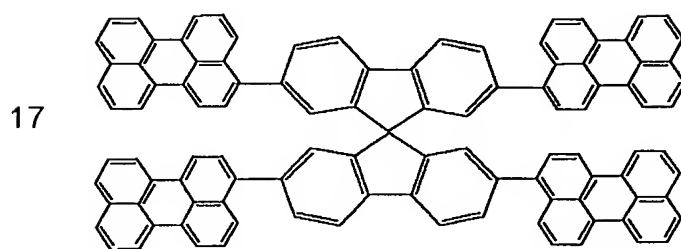
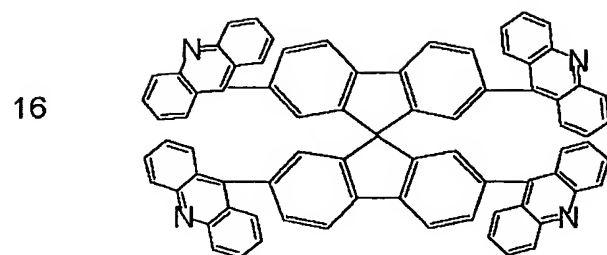
The substituents which the above-mentioned substituents may have include alkyl groups such as a methyl group, an ethyl group, and a propyl group; aralkyl groups such as a benzyl group, and a
10 phenethyl group; aryl groups such as a phenyl group, and a biphenyl group; heterocyclic groups such as a thienyl group, a pyrrolyl group, and a pyridyl group; amino groups such as a dimethylamino group, a diethylamino group, a dibenzylamino group, a
15 diphenylamino group, a ditolylamino group, and a dianisolylamino group; alkoxyl groups such as a methoxyl group, an ethoxyl group, a propoxyl group, and a phenoxy group; a cyano group; halogen atoms such as fluorine, chlorine, bromine, and iodine; and
20 the like.

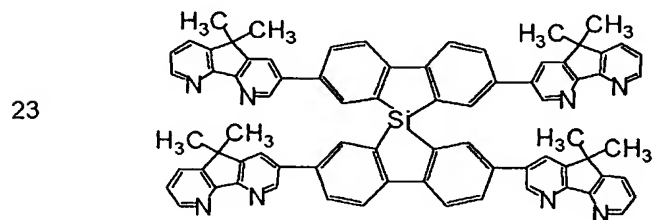
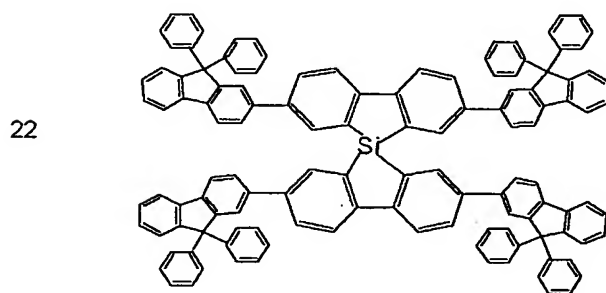
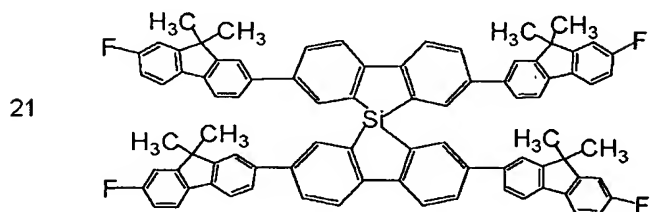
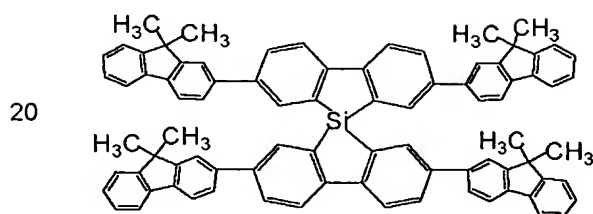
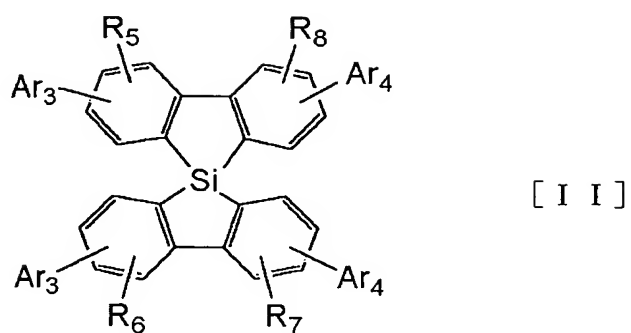
Next, although a typical example of the spiro compound of the present invention will be hereinafter given, the present invention is not limited thereto.

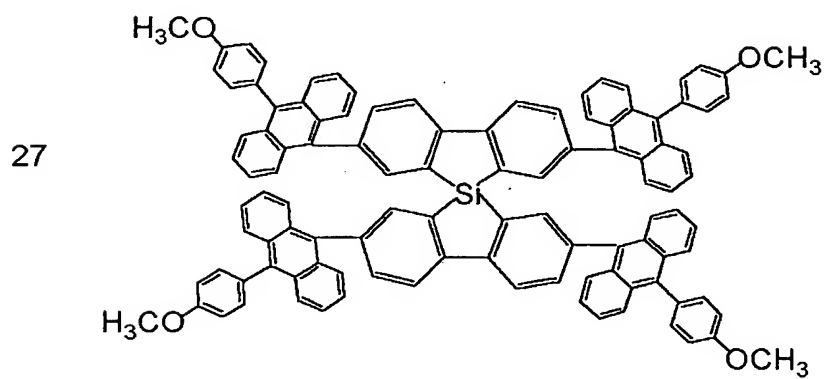
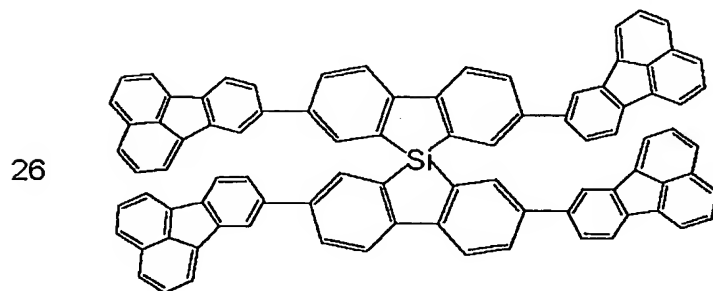
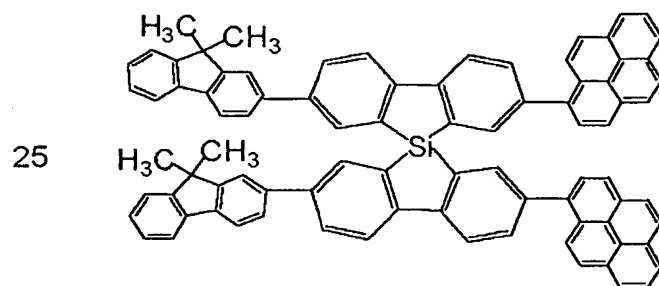
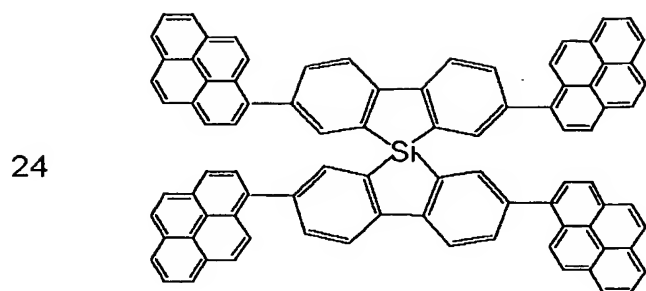


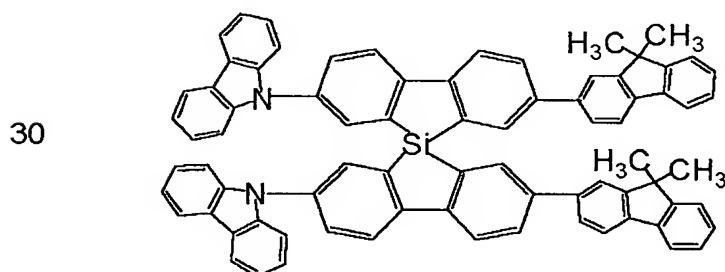
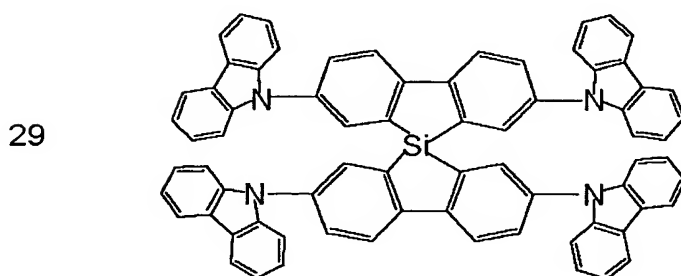
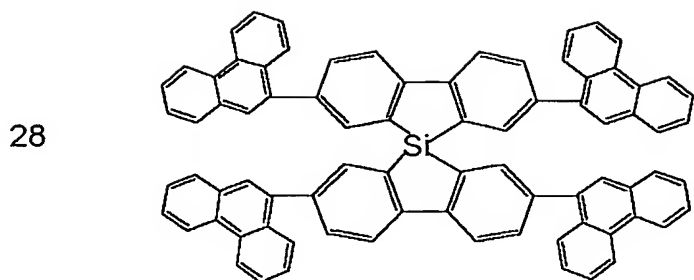












The spiro compound of the present invention can be synthesized by a generally well-known method, for example an intermediate of the spiro compound is

5 obtained by a method described in J. Org. Chem., 61, 6906, 1996, J. Am. Chem. Soc., 80, 1883, 1958, or the like, and furthermore the intermediate can be synthesized by the suzuki coupling method (e.g., Chem. Rev. 1995, 95, 2457-2483) using a palladium catalyst,

10 the Yamamoto method (e.g., Bull. Chem. Soc. Jpn. 51, 2091, 1978) using a nickel catalyst, a method in

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which a synthesis is performed by using a tin aryl compound (e.g., J. Org. Chem., 52, 4296, 1987), and so on.

As compared with the conventional compound, the
5 spiro compound of the present invention is a compound having excellent electron-transporting property, luminescence property and durability, which is useful for organic compound-containing layers of an organic luminescence device, particularly an electron-
10 transporting layer and a luminescent layer, and a layer formed by a vacuum evaporation method, a solution-coating method, or the like hardly causes crystallization or the like and is excellent in stability with time.

15 Next, the organic luminescence device of the present invention will be described in detail.

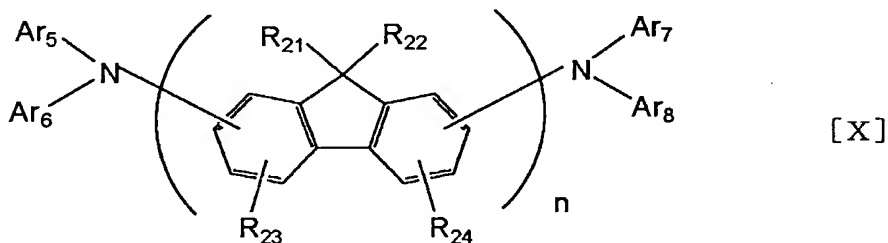
The organic luminescence device of the present invention includes at least a pair of electrodes including an anode and a cathode and one or plural
20 organic compound-containing layers sandwiched between the pair of electrodes, in which at least one layer of the organic compound-containing layers contains at least one spiro compound represented by the above general formula [I] or the general formula [II].

25 In the organic luminescence device of the present invention, it is preferable that at least an electron-transporting layer or a luminescent layer

among the organic-compound-containing layers contain at least one spiro compound.

In the organic luminescence device of the present invention, the spiro compound represented by the above general formula [I] or the general formula [II] is formed between the anode and the cathode by the vacuum evaporation method or the solution-coating method. The thickness of the organic layer is smaller than 10 μm , and it is preferable to make the layer as a thin film with a thickness of preferably 0.5 μm or less, more preferably 0.01 to 0.5 μm .

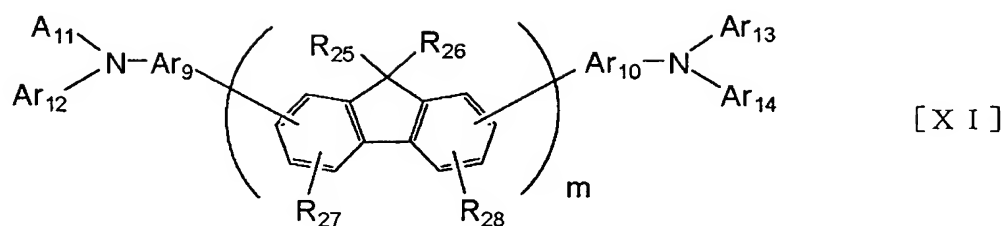
Further, according to a preferable mode of the organic luminescence device of the present invention, at least a luminescent layer among the layers containing the organic compound includes at least one spiro compound and a fluorene compound represented by one of the following general formula [X] and [XI]:



(where R_{21} and R_{22} represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, or a

- 22 -

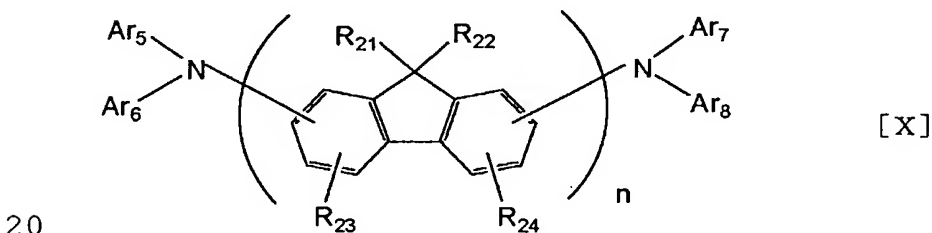
substituted or unsubstituted heterocyclic group, R_{21} themselves or R_{22} themselves, which are bonded to different fluorene groups, may be identical or different from each other, and R_{21} and R_{22} that are
5 bonded to the same fluorene group may be identical or different from each other; R_{23} and R_{24} represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a
10 substituted or unsubstituted heterocyclic group, a cyano group, or a halogen atom, and R_{23} themselves or R_{24} themselves, which are bonded to different fluorene groups, may be identical or different from each other, and R_{23} and R_{24} that are bonded to the same fluorene
15 group may be identical or different from each other; Ar_5 , Ar_6 , Ar_7 , and Ar_8 represent a substituted or unsubstituted aromatic group, a substituted or unsubstituted heterocyclic group, a substituted or unsubstituted condensed polycyclic aromatic group, or
20 a substituted or unsubstituted condensed polycyclic heterocyclic group, which may be identical or different from each other, and Ar_5 and Ar_6 as well as Ar_7 and Ar_8 may be bonded with each other to form rings, respectively; and n represents an integral
25 number of 1 to 10); and

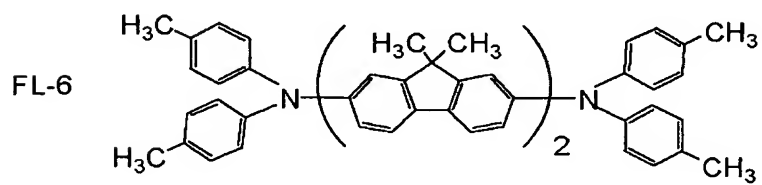
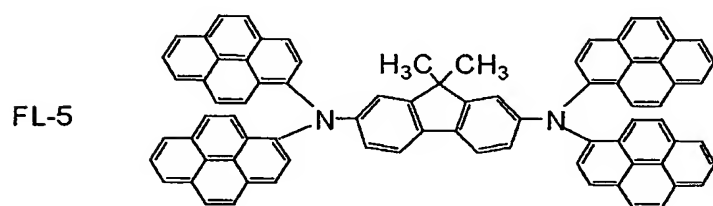
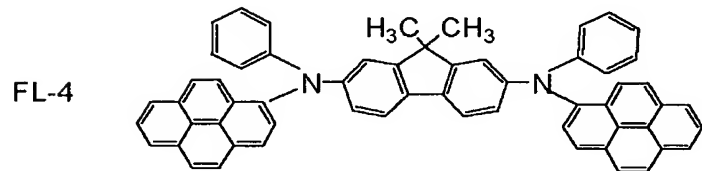
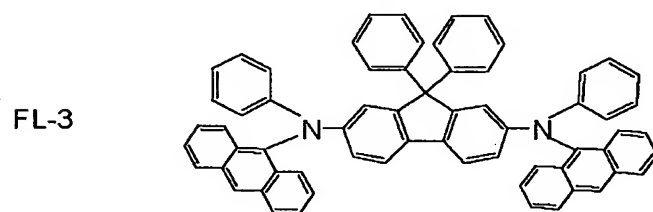
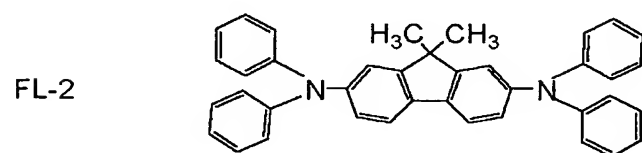
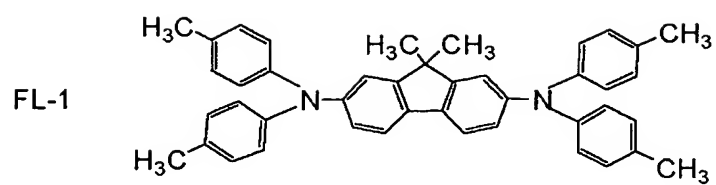


(where R_{25} and R_{26} represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, or a substituted or unsubstituted heterocyclic group, R_{25} themselves or R_{26} themselves, which are bonded to different fluorene groups, may be identical or different from each other, and R_{25} and R_{26} that are bonded to the same fluorene group may be identical or different from each other; R_{27} and R_{28} represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a cyano group, or a halogen atom, and R_{27} themselves or R_{28} themselves, which are bonded to different fluorene groups, may be identical or different from each other, and R_{27} and R_{28} that are bonded to the same fluorene group may be identical or different from each other; Ar_9 and Ar_{10} represent a substituted or unsubstituted divalent aromatic group or a substituted or unsubstituted divalent heterocyclic group, which may

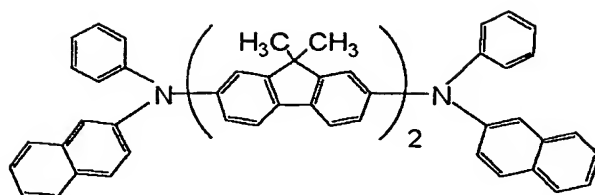
be identical or different from each other; Ar₁₁, Ar₁₂, Ar₁₃, and Ar₁₄ represent a substituted or unsubstituted aromatic group, a substituted or unsubstituted heterocyclic group, a substituted or unsubstituted condensed polycyclic aromatic group, or a substituted or unsubstituted condensed polycyclic heterocyclic group, which may be identical or different from each other, and Ar₁₁ and Ar₁₂ as well as Ar₁₃ and Ar₁₄ may be bonded with each other to form rings, respectively; and m represents an integral number of 1 to 10.)

Specific examples of substituents in the general formula [X] and [XI] are the same as those as in the cases of the above general formula [I] to [IX]. Typical examples of the fluorene compounds represented by the general formula [X] or [XI] will be given hereinafter, but the present invention is not limited thereto.

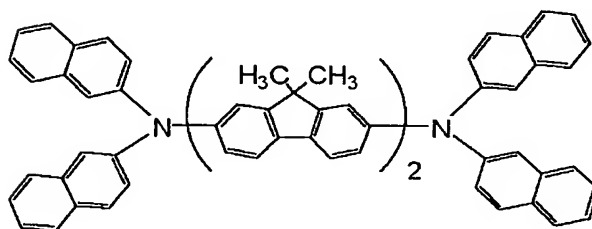




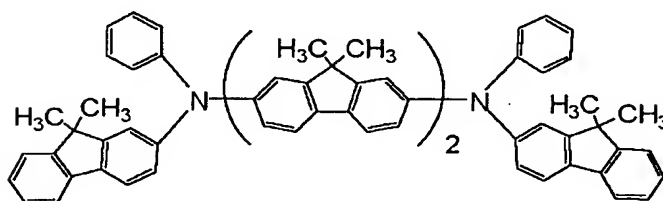
FL-7



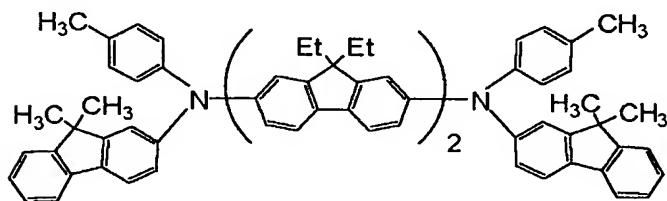
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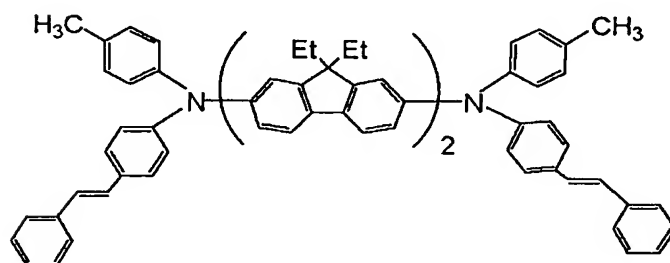
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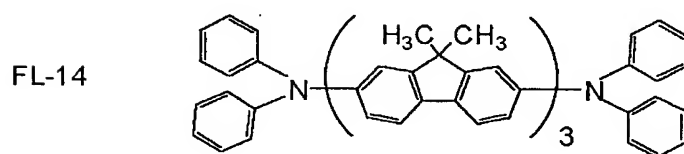
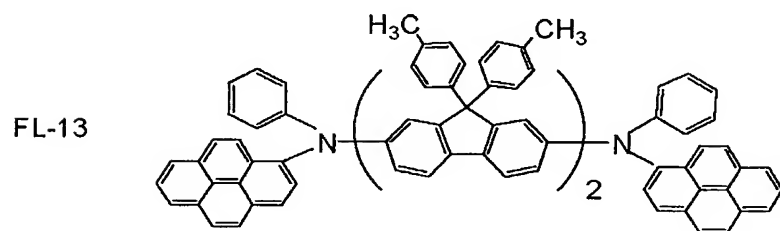
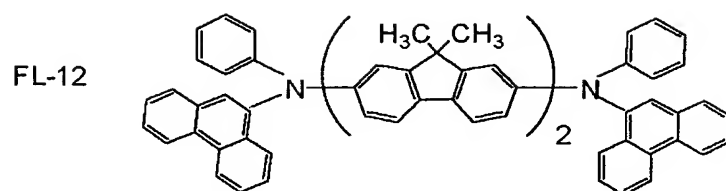


FL-10

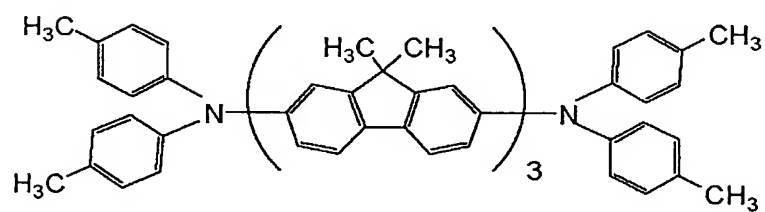


FL-11

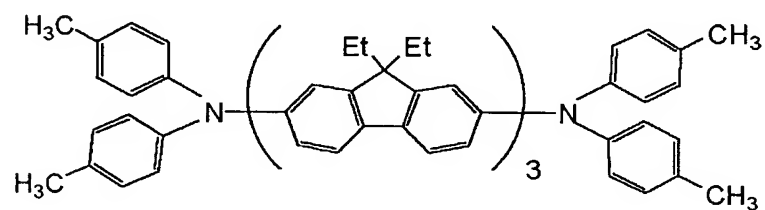


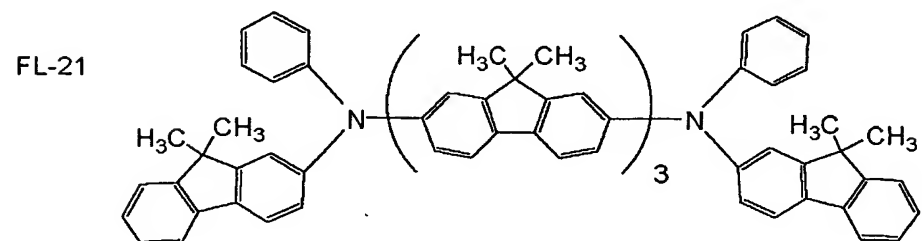
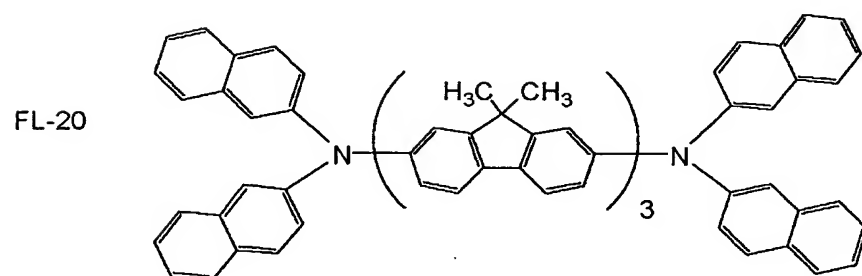
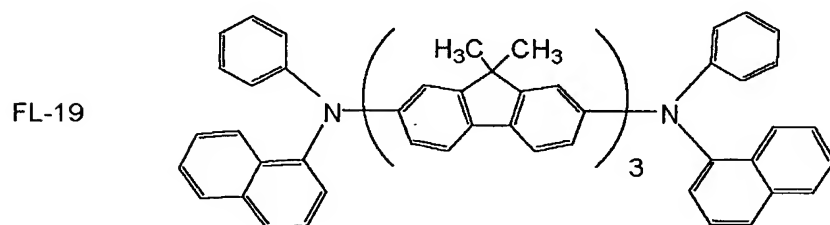
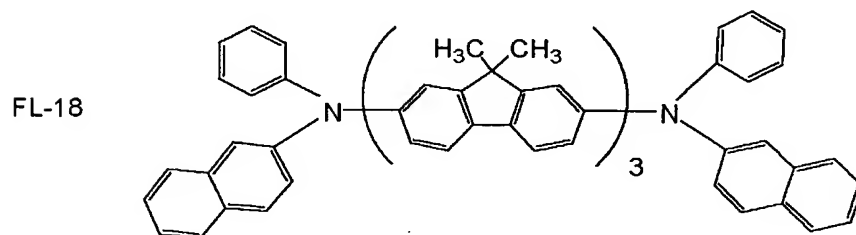
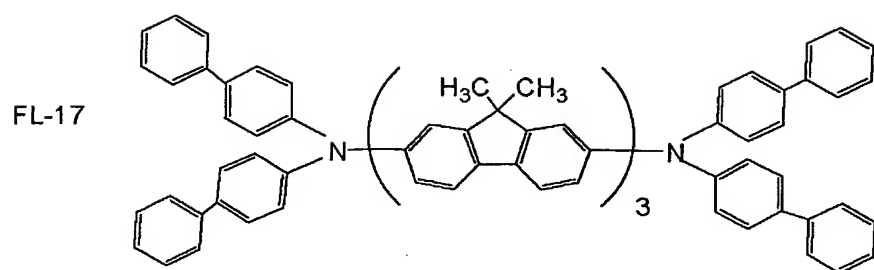


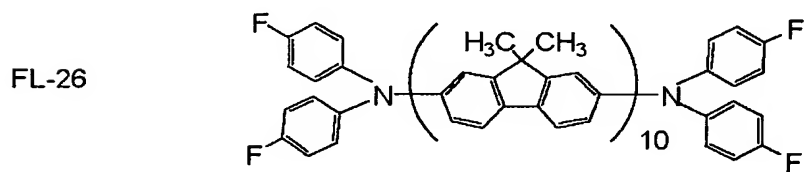
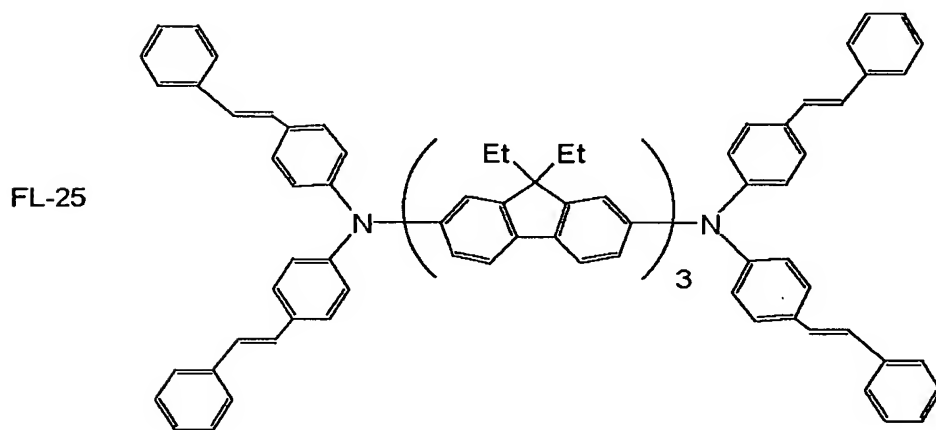
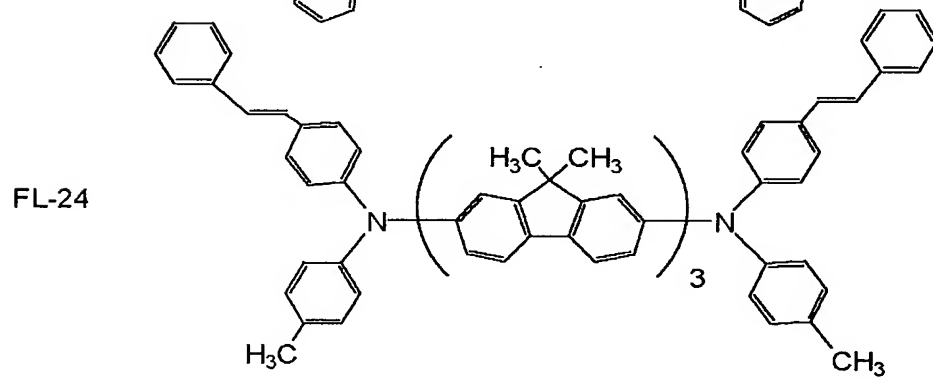
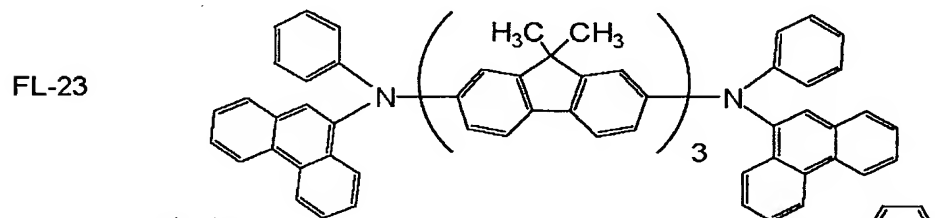
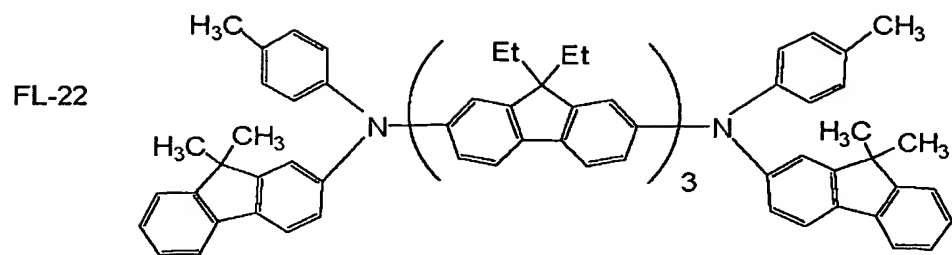
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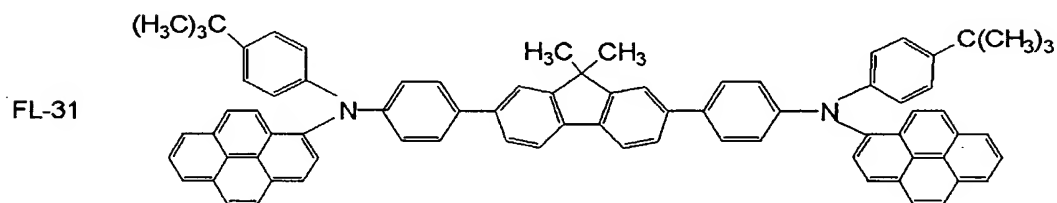
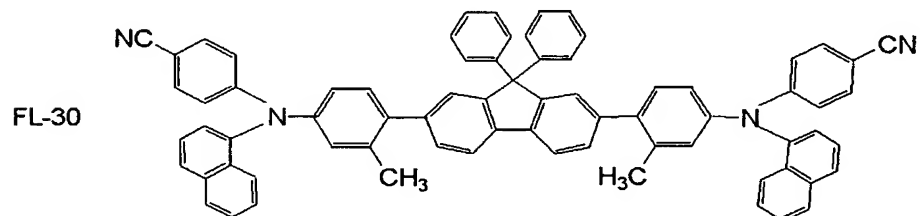
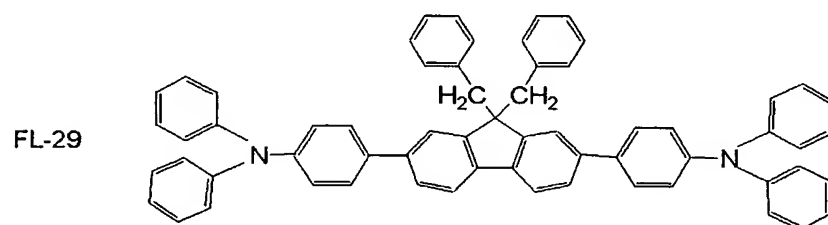
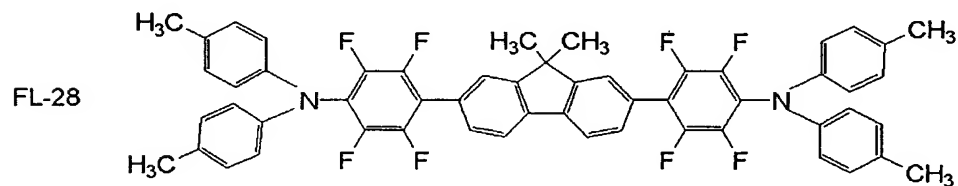
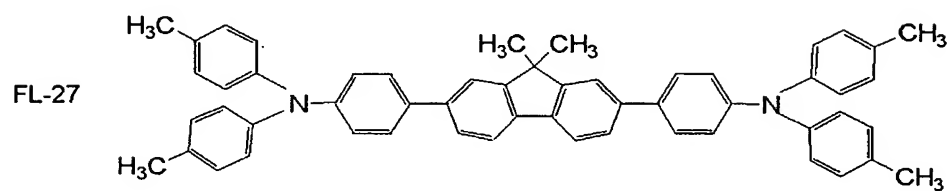
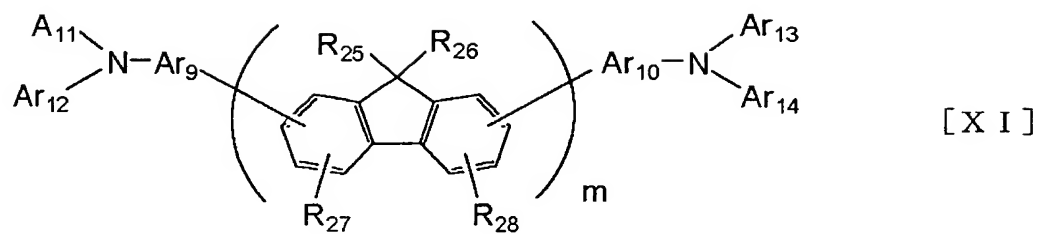


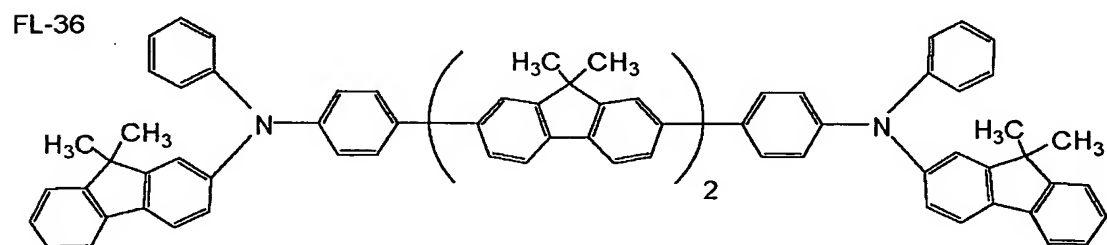
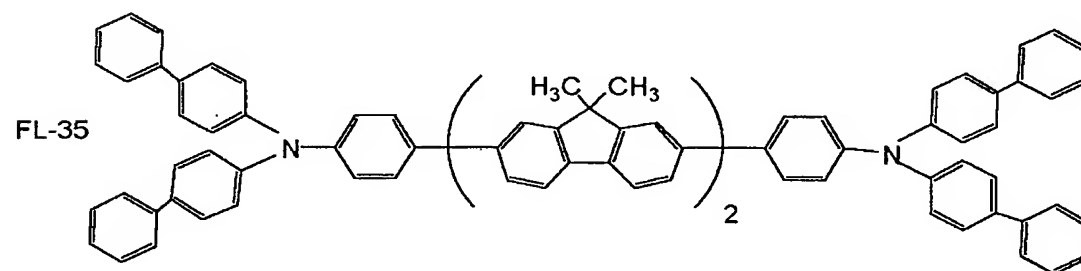
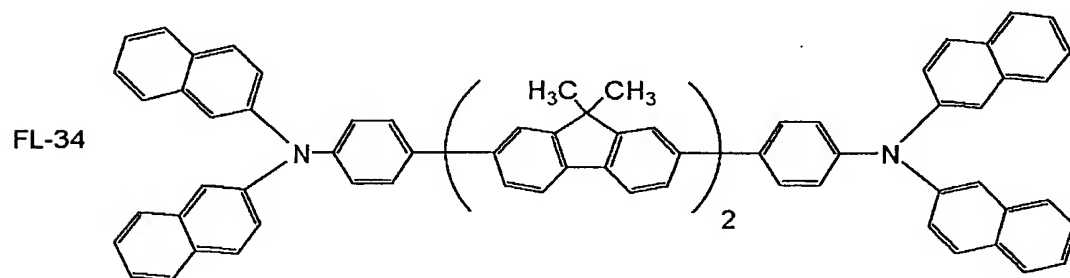
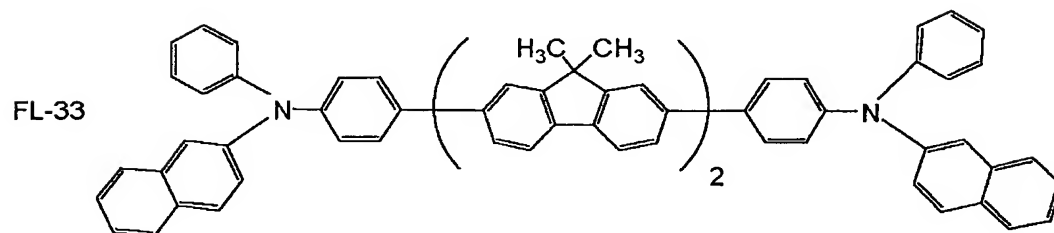
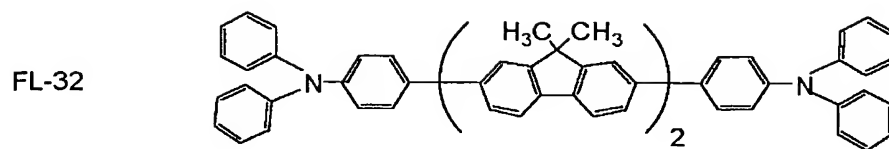
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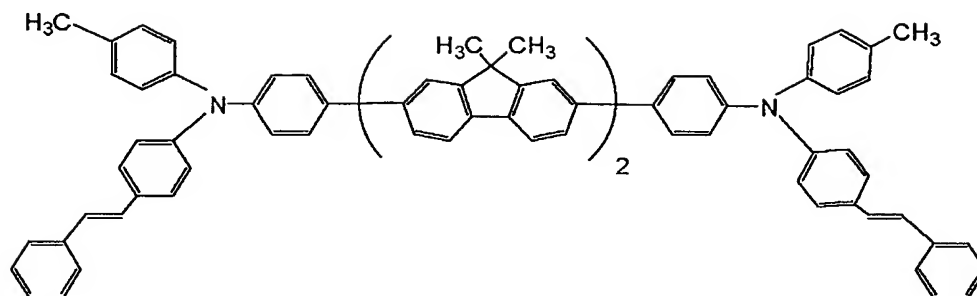




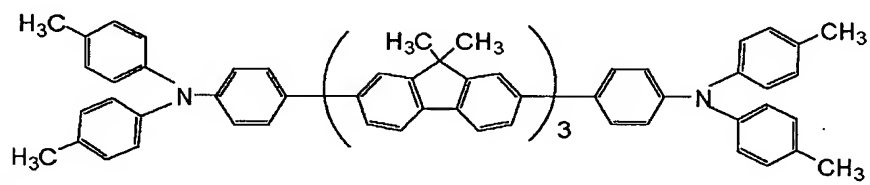




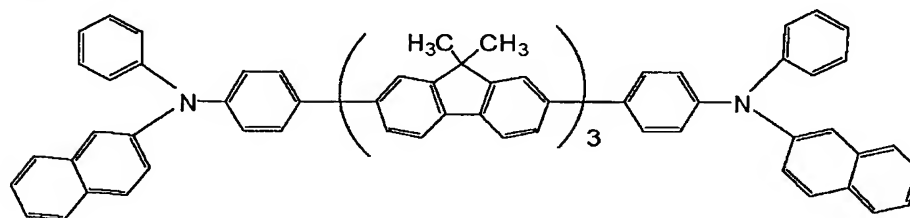
FL-37



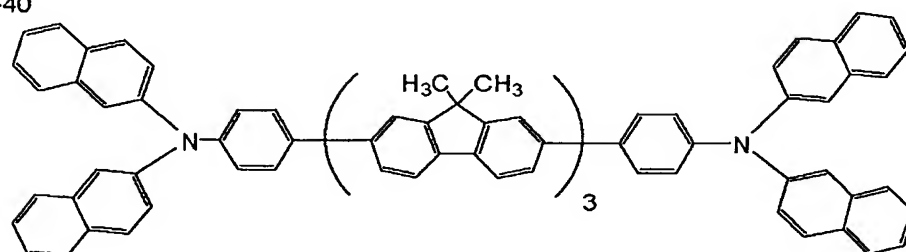
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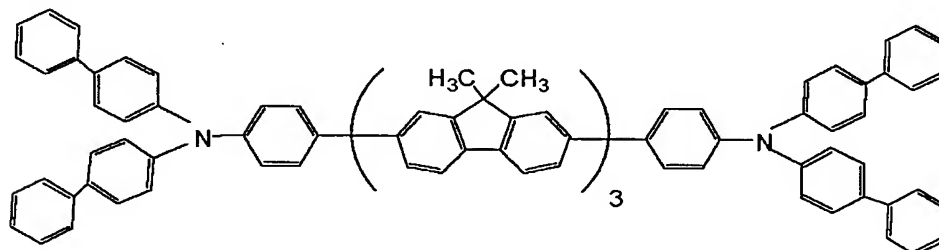
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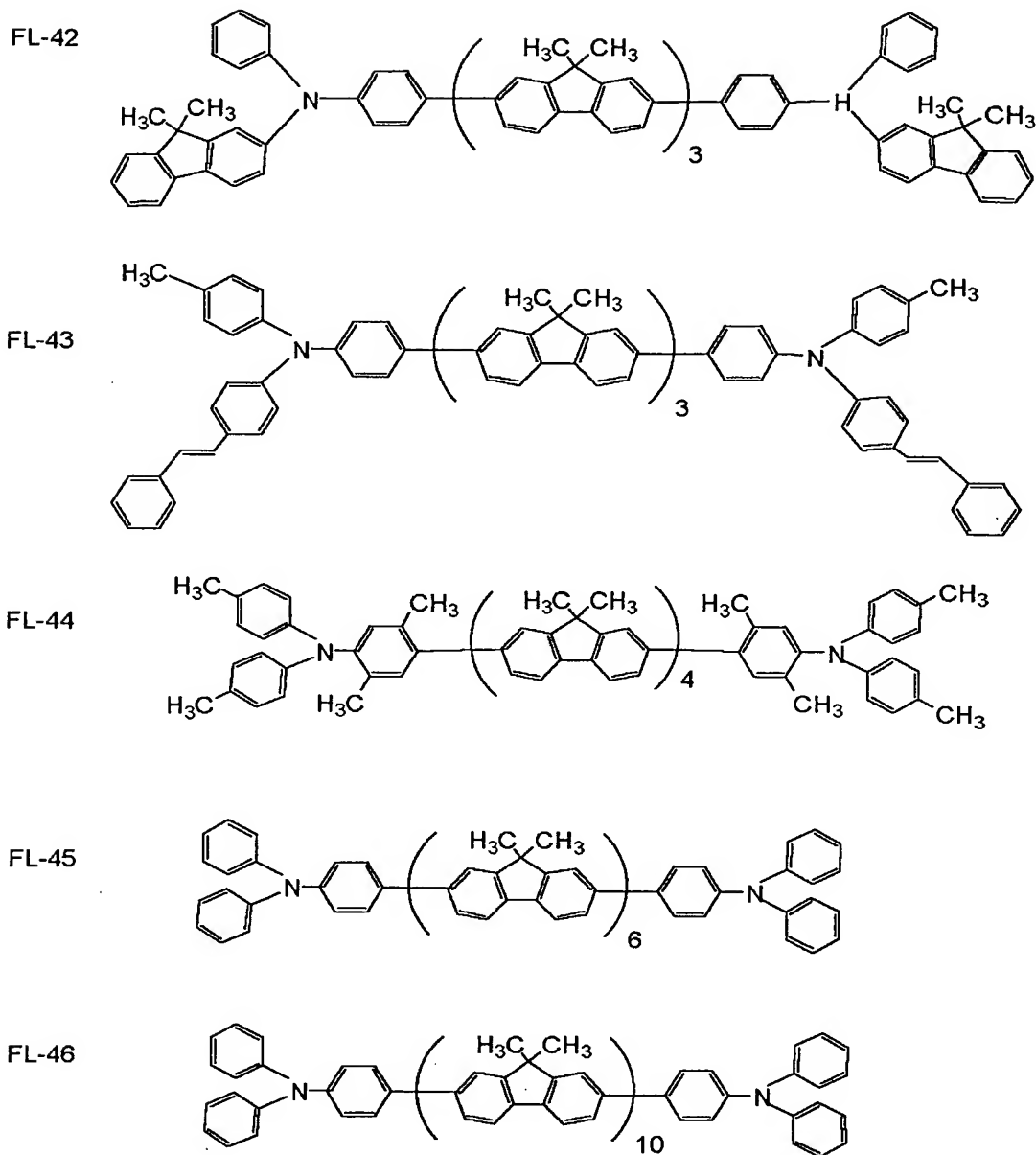


FL-40



FL-41





Preferable examples of the organic luminescence device are shown in Figs. 1 to 6, respectively.

5 Fig. 1 is a cross-sectional diagram that illustrates an example of the organic luminescence device of the present invention. In Fig. 1, the

device comprises an anode 2, a luminescent layer 3, and a cathode 4, which are formed on a substrate 1 in that order. The luminescence device used herein is useful when it singly has a hole-transporting ability, an electron-transporting ability, and a luminescence property in itself or when it is used in combination with compounds having those characteristics.

Fig. 2 is a cross-sectional diagram that illustrates another example of the organic luminescence device of the present invention. In Fig. 2, the device comprises an anode 2, a hole-transporting layer 5, an electron-transporting layer 6, and a cathode 4, which are formed on a substrate 1 in that order. In this case, a luminescent material is useful when a material having one or both of a hole-transporting property and an electron-transporting property is used for the respective layers and is used in combination with a hole-transporting material or an electron-transporting material having no luminescence property. In addition, in this case, the luminescent layer 3 is composed of either the hole-transporting layer 5 or the electron-transporting layer 6.

Fig. 3 is a cross-sectional diagram that illustrates another example of the organic luminescence device of the present invention. In Fig. 3, the device comprises an anode 2, a hole-

transporting layer 5, a luminescent layer 3, an
electron-transporting layer 6, and a cathode 4, which
are formed on a substrate 1 in that order. This is
one in which a carrier-transporting function and a
5 luminescence function are separated from each other,
and is used appropriately in combination with
compounds having a hole-transporting property, an
electron-transporting property, and a luminescence
property, respectively. Thus, the degree of freedom
10 in selecting a material increases extremely. In
addition, various kinds of compounds having different
luminescent wavelengths can be used. Therefore, the
diversity of luminescence hue can be allowed.
Furthermore, it also becomes possible to increase the
15 luminescence efficiency by effectively confining each
carrier or exciton in the middle luminescent layer 3.

Fig. 4 is a cross-sectional diagram that
illustrates another example of the organic
luminescence device of the present invention. In Fig.
20 4, as compared with the example of Fig. 3, the device
is constructed such that a hole-injection layer 7 is
inserted in the anode 2 side. It is effective in the
improvement of an adhesion between the anode 2 and
the hole-transporting layer 5 or the improvement of
25 an injection property of holes, so that it is
effective in lowering voltage.

Figs. 5 and 6 are cross-sectional diagrams that

illustrate other examples of the organic luminescence device of the present invention, respectively. In Figs. 5 and 6, as compared with the examples of Figs. 3 and 4, the device is constructed such that a layer (a hole-blocking layer 8) that prevents a hole or an exciton from passing toward the cathode 4 side is inserted between the luminescent layer 3 and the electron-transporting layer 6. The use of a compound having an extremely high ionization-potential as the hole-blocking layer 8 allows a configuration effective to an improvement in luminescence efficiency.

However, in Figs. 1 to 6, there are shown common basic device configurations. The configuration of the organic luminescence device using the compound of the present invention is not limited thereto. For instance, it is possible to adopt various layer configurations such as one in which an insulating layer is formed at the interface between the electrode and the organic layer, one in which an adhesive layer or an interference layer is formed, and one in which the hole-transporting layer is composed of two layers with different ionization potentials.

The spiro compounds represented by the general formula [I] or the general formula [II] to be used in the present invention are compounds each having

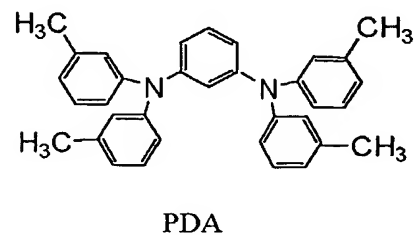
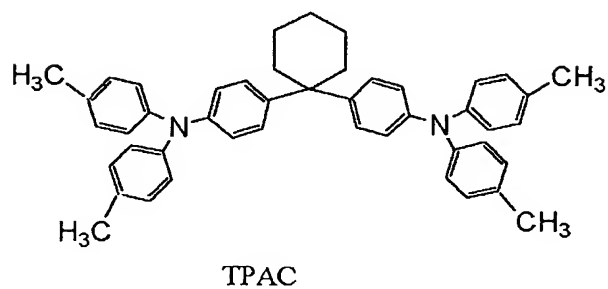
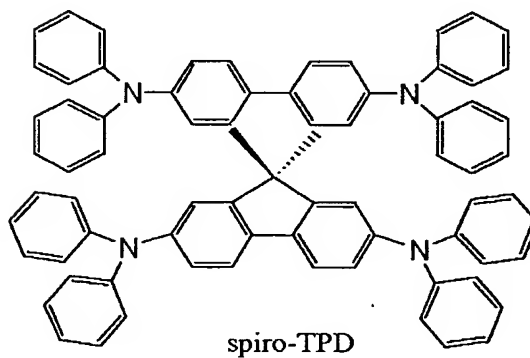
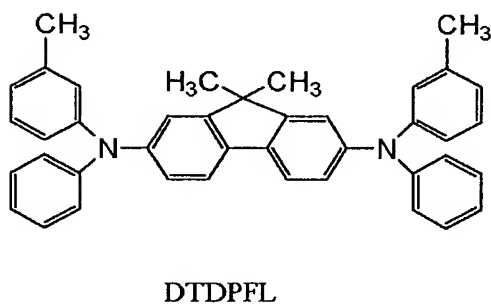
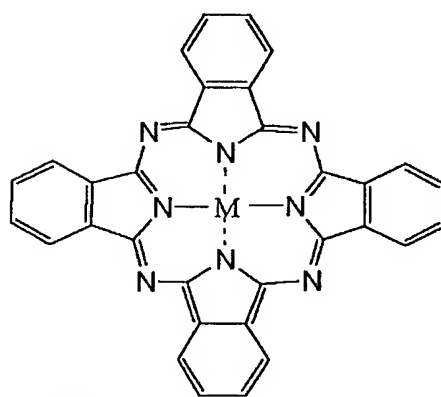
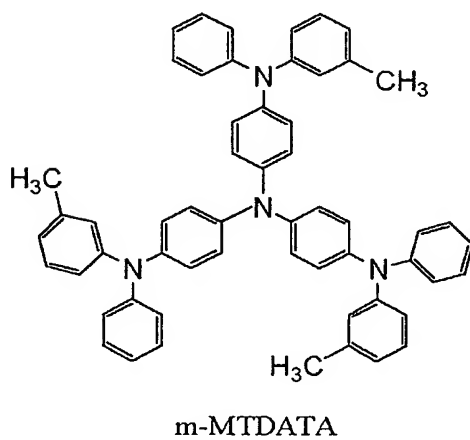
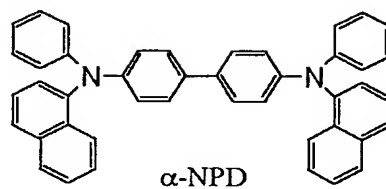
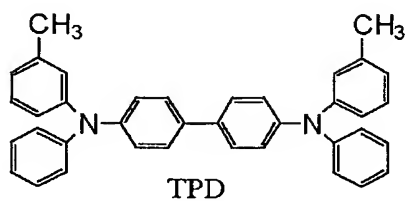
excellent electron-transporting property,
luminescence property, and durability as compared
with the conventional compounds, and they can be used
in any modes of Figs. 1 to 6.

5 In the present invention, the spiro compounds
represented by the general formula [I] or the general
formula [II] are used as structural components of the
electron-transporting layer or the luminescent layer.
However, hole-transporting compounds, luminescent
10 compounds, electron-transporting compounds, or the
like, which have been known, may be used together if
required.

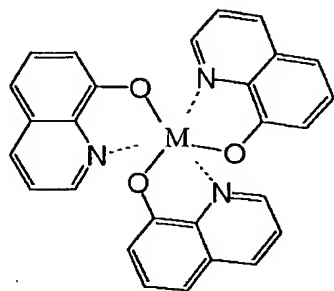
The examples of those compounds will be given
below.

15

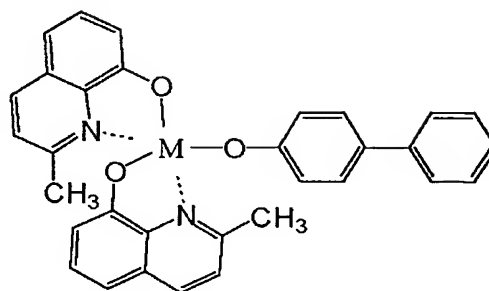
Hole-transporting compound



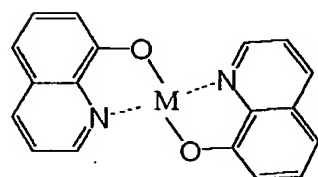
Electron-transporting luminescent material



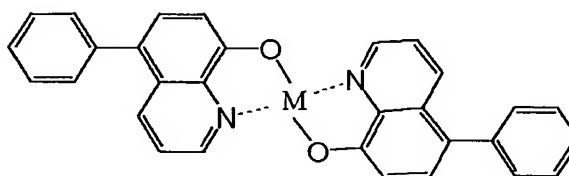
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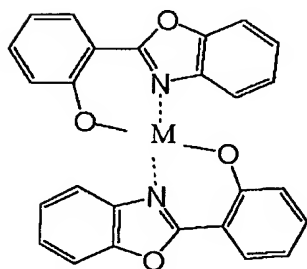
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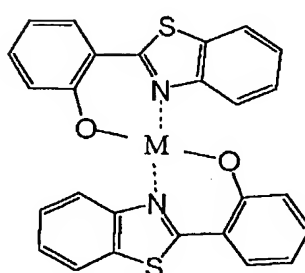
M : Zn , Mg , Be



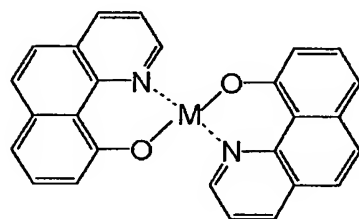
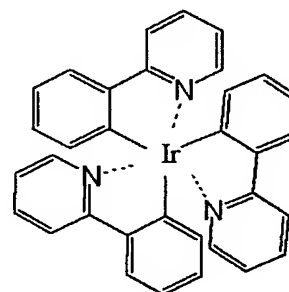
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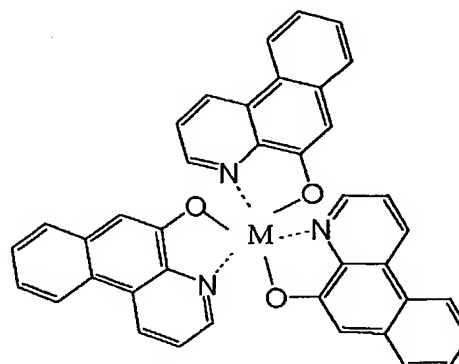
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M : Zn , Mg , Be

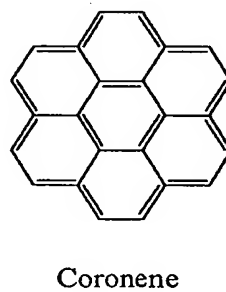
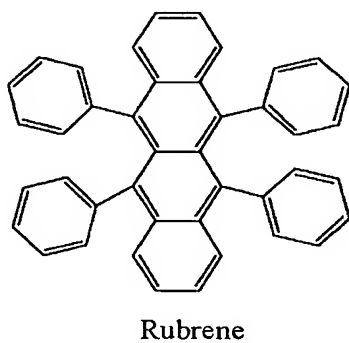
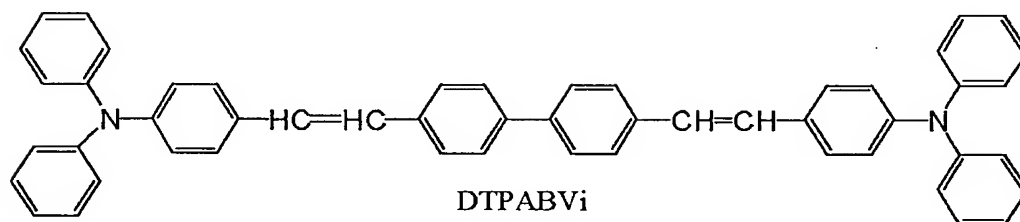
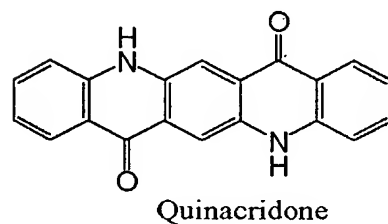
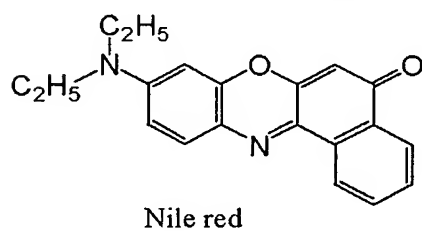
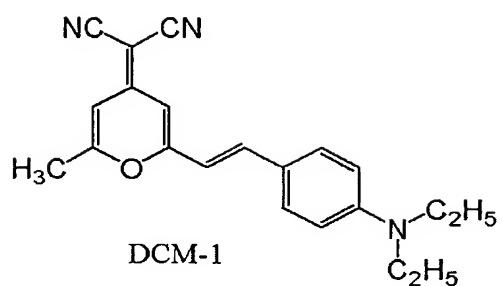
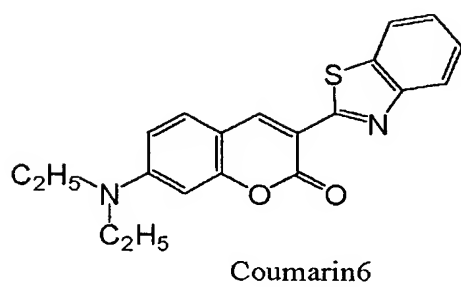


M : Zn , Mg , Be

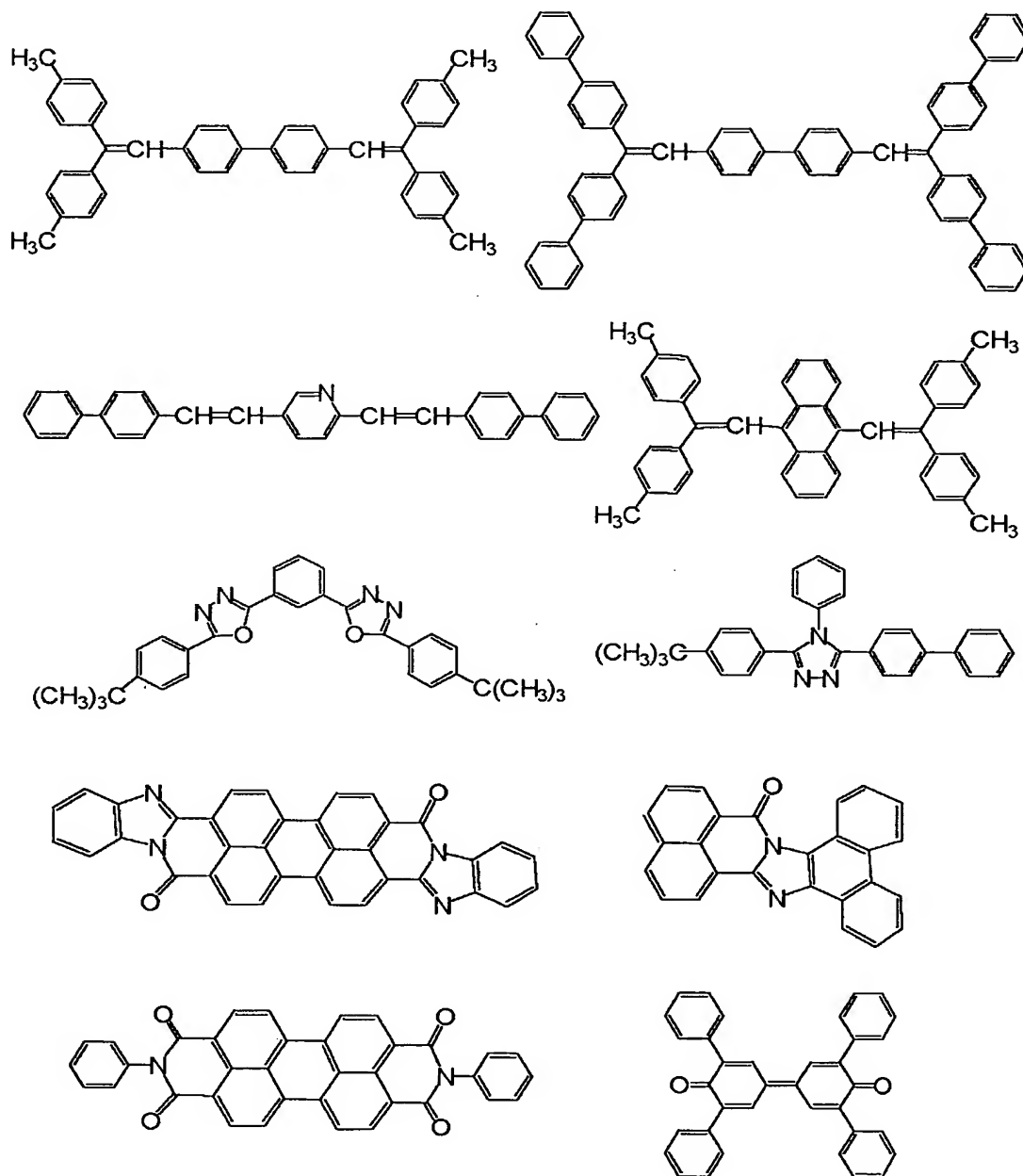


M : Al , Ga

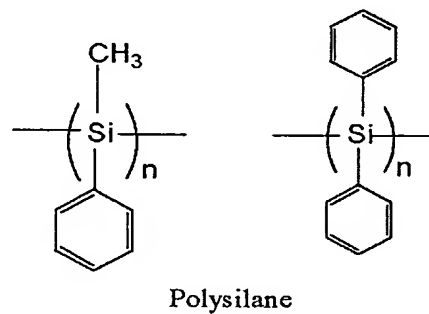
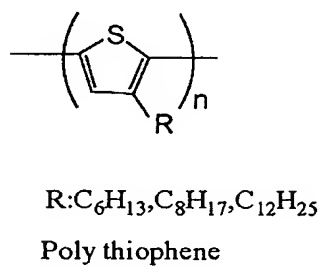
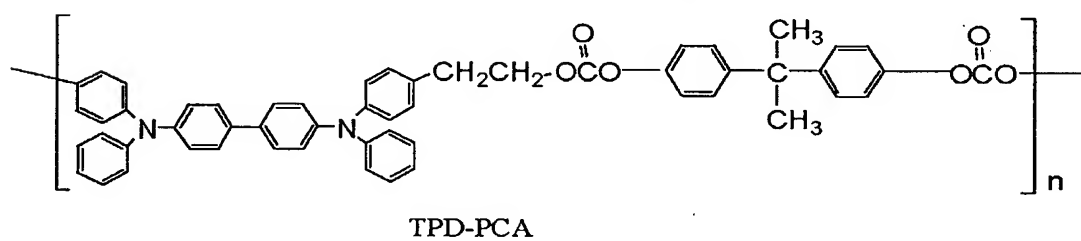
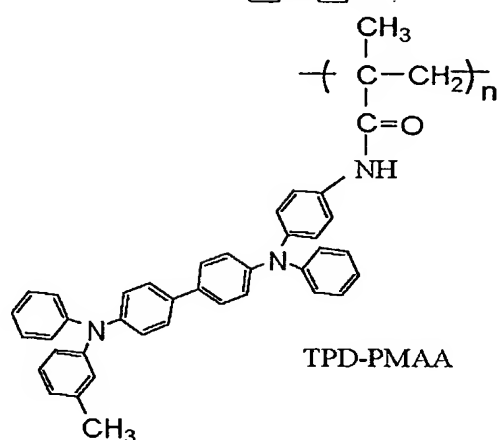
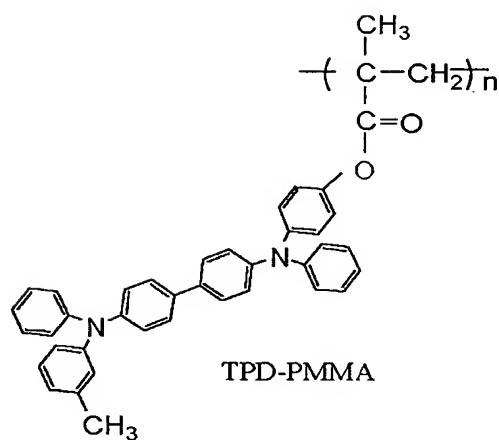
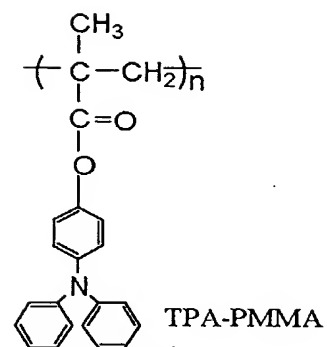
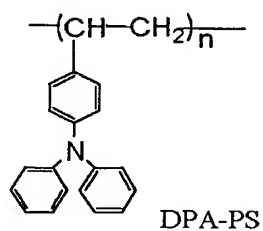
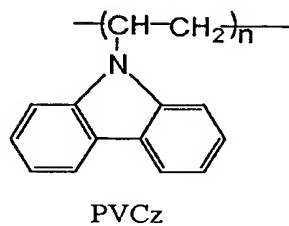
Luminescent material



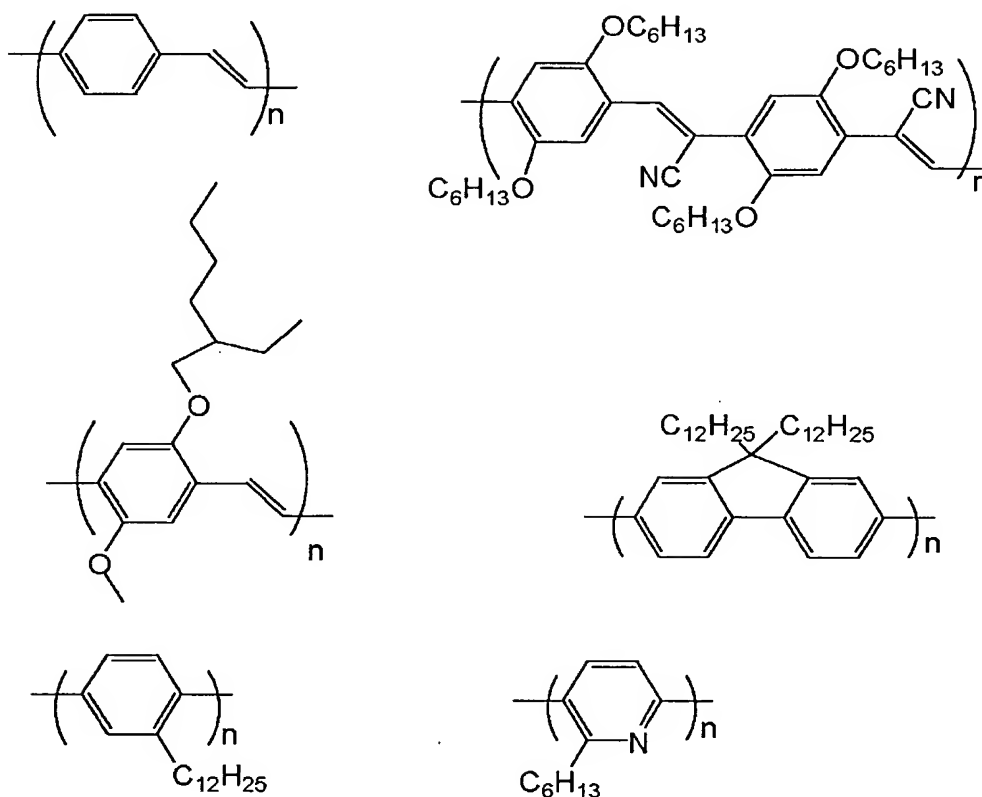
Luminescent layer matrix material and electron-transporting material



Polymeric hole-transporting material



Polymeric luminescent material and charge-
transporting material



5 In the organic luminescence device of the present invention, the layer containing the spiro compound represented by the general formula [I] or the general formula [II] and the layer containing another organic compound are generally formed as thin
10 films by a vacuum evaporation method, or by a coating method after being dissolved in an appropriate solvent. In particular, in the case of forming a

film with a coating method, the film may be formed in combination with an appropriate binder resin.

The above binder resin can be selected from a wide variety of the binder resins including, for
5 example, polyvinyl carbazole resin, polycarbonate resin, polyester resin, polyarylate resin, polystyrene resin, acrylic resin, methacryl resin, butyral resin, polyvinyl acetal resin, diallyl phthalate resin, phenol resin, epoxy resin, silicone
10 resin, polysulfone resin, and urea resin, although not limited to them. In addition, those resins may be used solely or one or more resins may be combined with each other as a copolymer.

The anode material may be one preferably having
15 a large work function. For example, a simple metal substance such as gold, platinum, nickel, palladium, cobalt, selenium, or vanadium, or an alloy thereof, or a metal oxide such as tin oxide, zinc oxide, indium tin oxide (ITO), or indium zinc oxide can be
20 used. In addition, a conductive polymer such as polyaniline, polypyrrole, polythiophene, or polyphenylene sulfide can be also used. Those electrode substances may be used solely or two or more substances may be used together.

25 On the other hand, the cathode material may be one preferably having a small work function. For example, a simple metal substance such as lithium,

sodium, potassium, cesium, calcium, magnesium, aluminum, indium, silver, lead, tin, or chromium, or an alloy of plural substances can be used. It is also possible to use a metal oxide such as indium tin
5 oxide (ITO). In addition, the cathode may be constructed as a single layer or may have a multi-layer configuration.

The substance used in the present invention may be, although not particularly limited to, a non-
10 transparent substrate such as a metallic substrate or a ceramic substrate, or a transparent substrate formed of glass, quartz, plastic sheets, or the like. In addition, it is also possible to control the luminescence color light by using a color filter film,
15 a fluorescent color-converting filter film, or a dielectric reflection film as a substrate.

Furthermore, a protective layer or a sealing layer may be formed on the prepared device for preventing the device from contacting with oxygen,
20 moisture, or the like. The protective layer may be a diamond thin film, a film made of an inorganic material such as a metal oxide or a metal nitride, or a polymer film made of a fluorocarbon resin, polyparaxylene, polyethylene, a silicone resin, or a
25 polystyrene resin, or furthermore it may be a photo-curing resin. Furthermore, it is also possible to package the device itself with an appropriate sealing

resin while covering it with a glass, a gas-impermeable film, a metal, or the like.

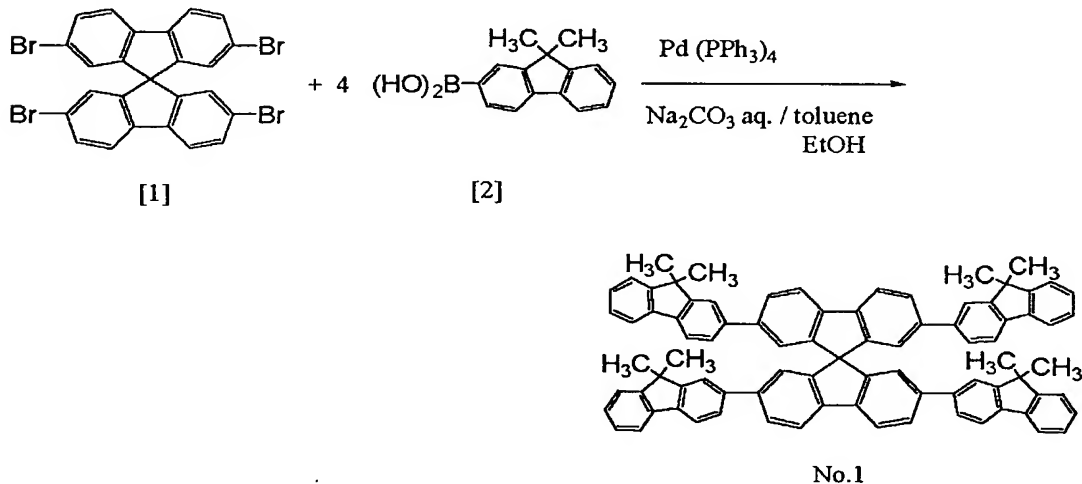
<Examples>

Hereinafter, the present invention will be described more specifically with examples. However, the present invention is not limited to those examples.

<Synthesis Example 1>

[Synthesis of Exemplified Compound No. 1]

10



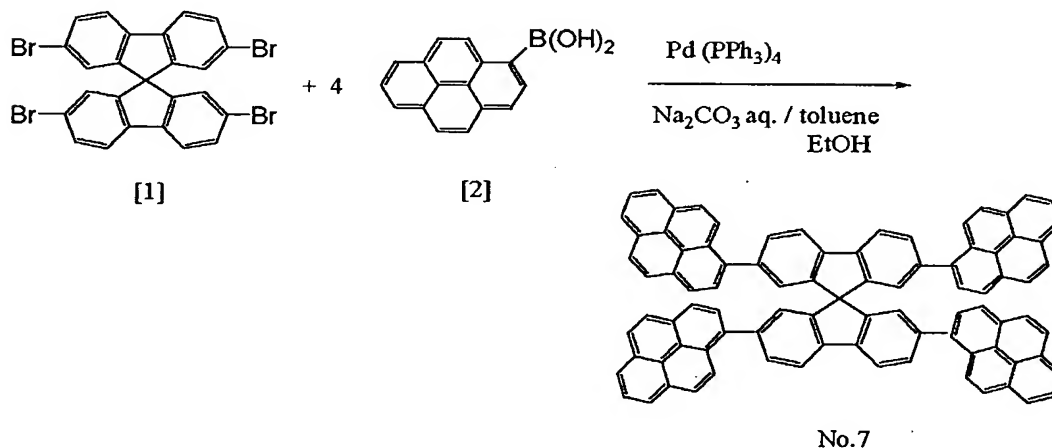
*1) J. Org. chem., 61, 6906-6921, 1996.

In a 500-ml three-neck flask, 2.0 g (3.16 mmol) of 2,2'-7,7'-tetrabromo-9,9'-spirobifluorene [1]^{*1)},
 15 4.5 g (19.0 mmol) of 9,9-dimethylfluorene-2-boronic acid [2], 140 ml of toluene, and 70 ml of ethanol were added, and an aqueous solution of 25 g of sodium carbonate / 130 ml of water was dropped thereto with stirring in a nitrogen atmosphere at room temperature,

followed by the addition of 0.7 g (0.63 mmol) of tetrakis (triphenylphosphine) palladium (0). After stirring the mixture for 30 minutes at room temperature, the temperature is allowed to rise to 77°C, followed by stirring for 8 hours. After the reaction, an organic layer was extracted with chloroform and was then dried with anhydrous sodium sulfate, followed by purification with a silica gel column (hexane + toluene mixture developing solvent). Consequently, 2.5 g (73% yield) of the exemplified compound No. 1 (white crystal) was obtained.

<Synthesis Example 2>

[Synthesis of Exemplified Compound No. 7]



15

In a 500-ml three-neck flask, 2.0 g (3.16 mmol) of 2,2'-7,7'-tetrabromo-9,9'-spirobifluorene [1], 4.7 g (19.0 mmol) of pyrene-1-boronic acid [2], 140 ml of toluene, and 70 ml of ethanol were added, and an

20

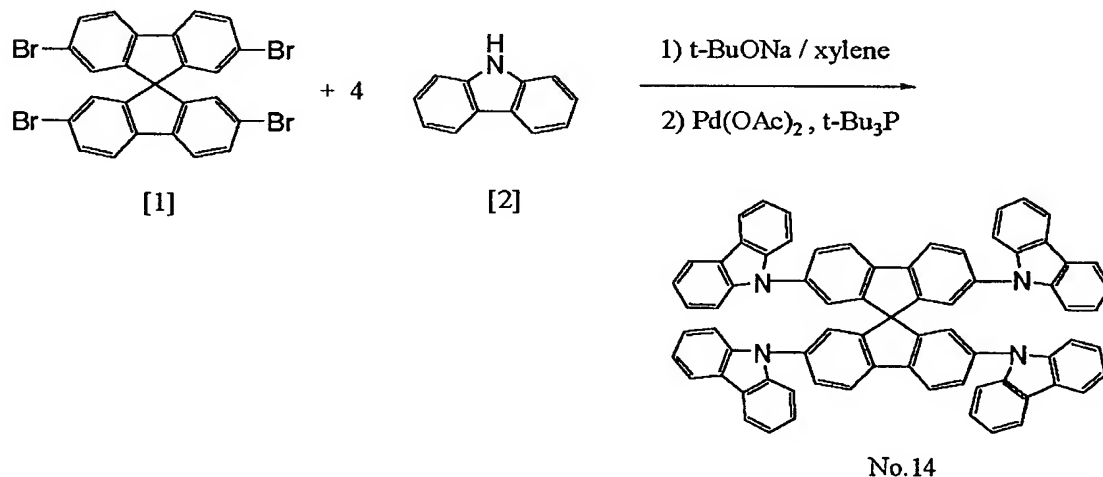
aqueous solution of 25 g of sodium carbonate / 130 ml of water was dropped with stirring in a nitrogen atmosphere at room temperature, followed by the addition of 0.7 g (0.63 mmol) of tetrakis

5 (triphenylphosphine) palladium (0). After stirring the mixture for 30 minutes at room temperature, the temperature is allowed to rise to 77°C, followed by stirring for 8 hours. After the reaction, an organic layer was extracted with chloroform and was then

10 dried with anhydrous sodium sulfate, followed by purification with a silica gel column (hexane + toluene mixture developing solvent). Consequently, 2.3 g (65% yield) of the exemplified compound No. 7 (white crystal) was obtained.

15 <Synthesis Example 3>

[Synthesis of Exemplified Compound No. 14]

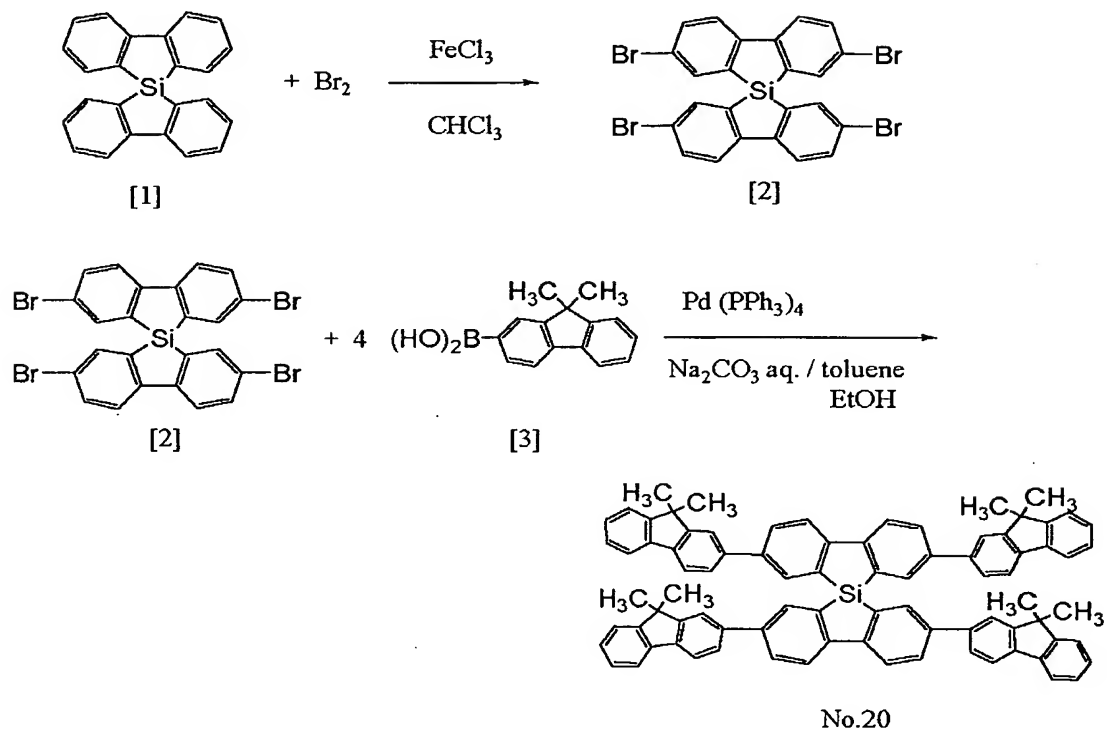


In a 300-ml three-neck flask, 2.0 g (3.16 mmol) of 2,2'-7,7'-tetrabromo-9,9'-spirobifluorene [1], 3.2

g (19.0 mmol) of carbazole [2], and 150 ml of xylene were added, and 2.0 g (20.9 mmol) of t-butoxy sodium was added thereto with stirring in a nitrogen atmosphere at room temperature, followed by heating
5 the mixture to a temperature of 50°C. In this mixture, a 5-ml xylene solution of 0.035 g (0.16 mmol) of palladium acetate and 0.032 g (0.16 mmol) of tri-t-butylphosphine was added, followed by heating the mixture to 130°C and stirring for 8 hours. After
10 the reaction, an organic layer was extracted with chloroform and was then dried with anhydrous sodium sulfate, followed by purification with a silica gel column (hexane + toluene mixture developing solvent). Consequently, 1.7 g (55% yield) of the exemplified
15 compound No. 14 (white crystal) was obtained.

<Synthesis Example 4>

[Synthesis of Exemplified Compound No. 20]



*2) J. Am. Chem. Soc., 80, 1883-1886, 1958.

- 5 In a 300-ml three-neck flask, 3.0 g (9.02 mmol) of 5,5'-spirobi(dibenzosilole) [1]^{*2)} and 100 ml of chloroform were added, and then 0.07 g (0.45 mmol) of iron chloride (III) was added with stirring at 0°C, followed by dropping 5.9 g (37.0 mmol) of bromine.
- 10 After stirring the mixture for 6 hours at room temperature, an organic layer was extracted with chloroform and was then washed with a sodium thiosulfate aqueous solution, followed by drying with anhydrous sodium sulfate. A crystal obtained by

distilling off the solvent was re-crystallized with chloroform, resulting in 4.0 g (69% yield) of tetrabromo-5,5'-spiro(dibenzosilole) [2] (white crystal).

5 Next, in a 500-ml three-neck flask, 2.0 g (3.09 mmol) of [2], 4.4 g (18.5 mmol) of 9,9-dimethylfluorene-2-boronic acid [3], 140 ml of toluene, and 70 ml of ethanol were added, and an aqueous solution of 25 g of sodium carbonate / 130 ml
10 of water was dropped in a nitrogen atmosphere, followed by the addition of 0.5 g (0.43 mmol) of tetrakis (triphenylphosphine) palladium (0). After stirring the mixture for 30 minutes at room temperature, the temperature is allowed to rise to
15 77°C, followed by stirring for 8 hours. After the reaction, an organic layer was extracted with chloroform and was then dried with anhydrous sodium sulfate, followed by purification with a silica gel column (hexane + toluene mixture developing solvent).
20 Consequently, 2.2 g (64% yield) of the exemplified compound No. 20 (white crystal) was obtained.

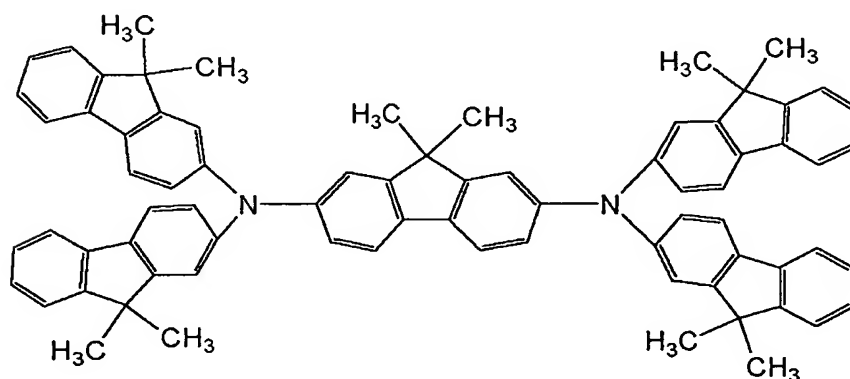
<Example 1>

A device having the structure shown in Fig. 2 was prepared.

25 On a glass substrate as a substrate 1, indium tin oxide (ITO) is deposited into a film with a thickness of 120 nm by a sputtering method to obtain

an anode 2, so that the substrate thus formed is used as a transparent conductive support substrate. This was sequentially subjected to ultrasonic cleanings with acetone and isopropyl alcohol (IPA), and was
5 then washed with IPA by boiling, followed by drying. Furthermore, one subjected to UV/ozone cleaning was used as a transparent conductive support substrate.

On the transparent conductive support substrate, a chloroform solution of the compound represented by
10 the following structural formula was coated into a film of 30 nm in thickness by a spin-coating method, resulting in a hole-transporting layer 5.



Furthermore, an electron-transporting layer 6
15 was formed by forming a film of 50 nm in thickness from a spiro compound represented by the exemplified compound No. 1 by a vacuum evaporation method. The film formation was performed under the conditions in which the degree of vacuum at the time of evaporation
20 was 1.0×10^{-4} Pa and the film formation rate was 0.2 to 0.3 nm/sec.

A metal layer film of 50 nm in thickness was formed on the above organic layer as a cathode 4 by using an evaporation material including aluminum and lithium (lithium concentration: 1% by atom) by a vacuum evaporation method, and further an aluminum layer of 150 nm in thickness was formed by a vacuum evaporation method. The film formation was performed under the conditions in which the degree of vacuum at the time of evaporation was 1.0×10^{-4} Pa and the film formation rate was 1.0 to 1.2 nm/sec.

Furthermore, the resulting product was covered with a protective glass plate in a nitrogen atmosphere and was then sealed with an acrylic resin adhesive.

When a direct current voltage of 10 V was applied on the device obtained in this way with an ITO electrode (anode 2) provided as a positive electrode and an Al-Li electrode (cathode 4) provided as a negative electrode, the current was caused to flow into the device at a current density of 11.5 mA/cm² and blue-colored luminescence at a luminance of 3800 cd/m² was observed.

Furthermore, when the current density was kept at 10.0 mA/cm² and the voltage was applied for 100 hours, an initial luminance of 3500 cd/m² changed to a luminance of 3300 cd/m² after 100 hours, indicating small deterioration of luminance.

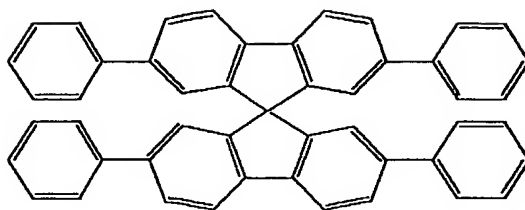
<Examples 2 to 10>

Devices were prepared and evaluated in the same way as that of Example 1, except that the exemplified compounds shown in Table 1 were used in place of the
5 exemplified compound No. 1. The results are shown in Table 1.

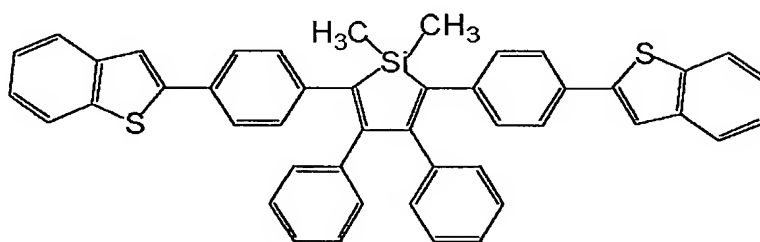
<Comparative Examples 1 to 3>

Devices were prepared and evaluated in the same way as that of Example 1, except that the compounds
10 represented by the following structural formula were used in place of the exemplified compound No. 1. The results are shown in Table 1.

Comparative Compound No. 1



Comparative Compound No. 2



Comparative Compound No. 3

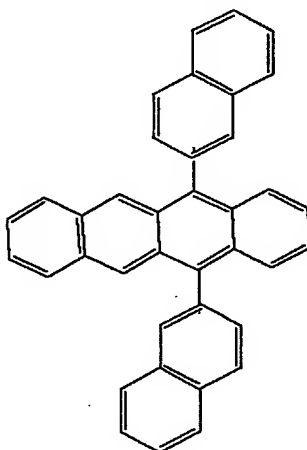


Table 1

Example No.	Exemplified compound No.	Initial stage		Current density (mA/cm ²)	Durability	
		Applied voltage (V)	Luminance (cd/m ²)		Initial luminance (cd/m ²)	Luminance after 100-hour (cd/m ²)
Example 1	1	10	3800	10.0	3500	3300
2	5	10	4500	10.0	4100	3800
3	7	10	3500	10.0	3300	3200
4	10	10	2900	10.0	2700	2500
5	13	10	3800	10.0	3500	3400
6	16	10	2400	10.0	2200	1900
7	18	10	2400	10.0	2300	2200
8	21	10	3600	10.0	3500	3300
9	24	10	2700	10.0	2600	2200
10	27	10	2300	10.0	2100	2000
Comparative Example 1	Comparative 1	10	850	10.0	800	550
2	Comparative 2	10	700	10.0	650	250
3	Comparative 3	10	400	10.0	350	100

<Example 11>

A device having the structure shown in Fig. 3 was prepared.

In the same manner as in Example 1, a hole-transporting layer 5 was formed on the transparent conductive support substrate.

Further, a luminescent layer 3 was formed by forming a film of 20 nm in thickness from a spiro compound represented by the exemplified compound No. 3 by a vacuum evaporation method. The film formation was performed under the conditions in which the degree of vacuum at the time of evaporation was 1.0×10^{-4} Pa and the film formation rate was 0.2 to 0.3 nm/sec.

Furthermore, an electron-transporting layer 6 was formed by forming a film of 40 nm in thickness from aluminum tris quinolinol by a vacuum evaporation method. The film formation was performed under the conditions in which the degree of vacuum at the time of evaporation was 1.0×10^{-4} Pa and the film formation rate was 0.2 to 0.3 nm/sec.

Next, after forming a cathode 4 in the same manner as in Example 1, the resulting product was sealed.

When a direct current voltage of 8 V was applied on the device obtained in this way with an ITO electrode (anode 2) provided as a positive

electrode and an Al-Li electrode (cathode 4) provided as a negative electrode, the current was caused to flow into the device at a current density of 12.0 mA/cm² and blue-colored luminescence at a luminance of 6700 cd/m² was observed.

Furthermore, when the current density was kept at 10.0 mA/cm² and the voltage was applied for 100 hours, an initial luminance of 5500 cd/m² changed to a luminance of 5200 cd/m² after 100 hours, indicating small deterioration of luminance.

<Examples 12 to 20>

Devices were prepared and evaluated in the same way as that of Example 11, except that the exemplified compounds shown in Table 2 were used in place of the exemplified compound No. 3. The results are shown in Table 2.

<Comparative Examples 4 to 6>

Devices were prepared and evaluated in the same way as that of Example 11, except that the comparative compounds No. 1 to No. 3 were used in place of the exemplified compound No. 3. The results are shown in Table 2.

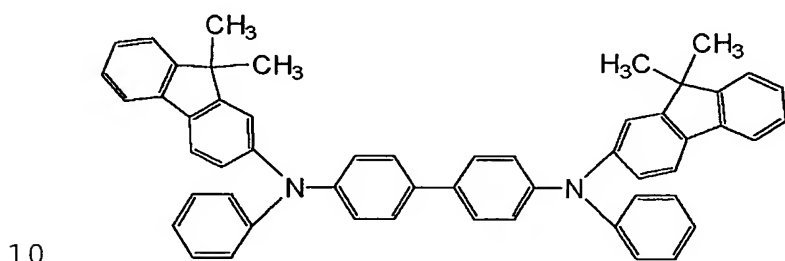
Table 2

Example No.	Exemplified compound No.	Initial stage		Durability		
		Applied voltage (V)	Luminance (cd/m ²)	Current density (mA/cm ²)	Initial luminance (cd/m ²)	Luminance after 100-hour (cd/m ²)
Example 11	3	8	6700	10.0	5500	5200
12	6	8	6500	10.0	5200	4800
13	8	8	7300	10.0	6000	5700
14	11	8	5200	10.0	4400	4100
15	15	8	3900	10.0	3600	3400
16	19	8	5100	10.0	4200	4000
17	23	8	5600	10.0	4600	4300
18	25	8	3600	10.0	2700	2500
19	26	8	3800	10.0	2900	2600
20	28	8	5200	10.0	4100	3800
Comparative Example 4	Comparative 1	8	800	10.0	770	450
5	Comparative 2	8	500	10.0	400	150
6	Comparative 3	8	1200	10.0	900	300

<Example 21>

A device having the structure shown in Fig. 3 was prepared.

On a transparent conductive support substrate similar to that in Example 1, a chloroform solution of a compound represented by the following structural formula was applied into a film of 20 nm in thickness by a spin-coating method, resulting in a hole-transporting layer 5.



Furthermore, the spiro compound represented as the exemplified compound No. 7 and the fluorene compound represented as the exemplified compound No. FL-1 (weight ratio of 100 : 1) were deposited into a film with a thickness of 20 nm by the vacuum evaporation method to form a luminescent layer 3. The film formation was performed under the conditions in which the degree of vacuum at the time of evaporation was 1.0×10^{-4} Pa and the film formation rate was 0.2 to 0.3 nm/sec.

Furthermore, an electron-transporting layer 6 was formed by forming a film of 40 nm in thickness from aluminum tris quinolinol by a vacuum evaporation

method. The film formation was performed under the conditions in which the degree of vacuum at the time of evaporation was 1.0×10^{-4} Pa and the film formation rate was 0.2 to 0.3 nm/sec.

5 Next, after forming a cathode 4 in the same manner as in Example 1, the resulting product was sealed.

 When a direct current voltage of 8 V was applied on the device obtained in this way with an
10 ITO electrode (anode 2) provided as a positive electrode and an Al-Li electrode (cathode 4) provided as a negative electrode, the current was caused to flow into the device at a current density of 13.5 mA/cm² and blue-colored luminescence at a luminance
15 of 16000 cd/m² was observed.

 Furthermore, when the current density was kept at 10.0 mA/cm² and the voltage was applied for 100 hours, the initial luminance of 12000 cd/m² changed to a luminance of 9000 cd/m² after 100 hours, indicating
20 small deterioration of luminance.

<Examples 22 to 53>

 Devices were prepared and evaluated in the same way as that of Example 21, except that the exemplified fluorene compound shown in Table 3 was
25 used in place of the exemplified fluorene compound No. FL-1. The results are shown in Table 3.

<Comparative Examples 7 to 9>

Devices were prepared and evaluated in the same way as that of Example 21, except that the comparative compounds No. 1 to No. 3 were used in place of the exemplified compound No. 7. The results
5 are shown in Table 3.

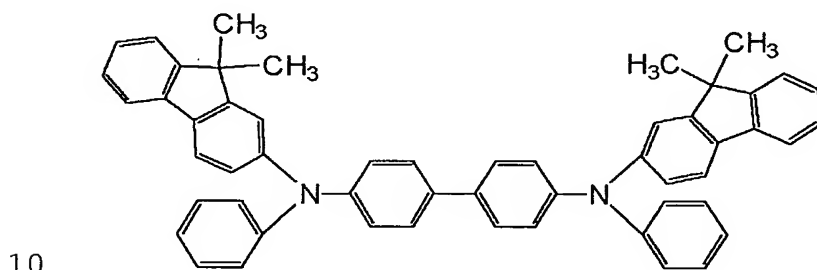
Table 3

Example No.	Exemplified compound No.	Exemplified fluorene compound No.	Initial stage		Durability		
			Applied voltage (V)	Luminance (cd/m ²)	Current density (mA/cm ²)	Initial luminance (cd/m ²)	Luminance after 100-hour (cd/m ²)
Example 21	7	FL-1	8	16000	10.0	12000	9000
22	7	FL-2	8	15000	10.0	12000	8000
23	7	FL-3	8	17000	10.0	14000	10000
24	7	FL-4	8	12000	10.0	8500	7000
25	7	FL-5	8	9000	10.0	7000	6000
26	7	FL-6	8	19000	10.0	14000	11000
27	7	FL-7	8	20000	10.0	14000	12000
28	7	FL-9	8	22000	10.0	17000	13000
29	7	FL-10	8	21000	10.0	16000	13000
30	7	FL-12	8	17000	10.0	11000	8500
31	7	FL-13	8	13000	10.0	10000	7000
32	7	FL-14	8	18000	10.0	16000	14000
33	7	FL-15	8	19000	10.0	16000	14000
34	7	FL-18	8	19000	10.0	17000	15000
35	7	FL-21	8	23000	10.0	19000	16000
36	7	FL-24	8	24000	10.0	19000	17000
37	7	FL-26	8	9500	10.0	8000	6000
38	7	FL-27	8	17000	10.0	13000	11000
39	7	FL-28	8	10000	10.0	8000	6500
40	7	FL-29	8	8000	10.0	7000	6000
41	7	FL-30	8	9500	10.0	8000	6500
42	7	FL-31	8	12000	10.0	10000	7000
43	7	FL-32	8	23000	10.0	18000	15000
44	7	FL-33	8	23000	10.0	17000	14000
45	7	FL-36	8	25000	10.0	19000	16000
46	7	FL-37	8	25000	10.0	18000	15000
47	7	FL-38	8	20000	10.0	17000	14000
48	7	FL-39	8	24000	10.0	19000	16000
49	7	FL-41	8	26000	10.0	20000	16000
50	7	FL-42	8	26000	10.0	21000	17000
51	7	FL-44	8	15000	10.0	12000	9500
52	7	FL-45	8	12000	10.0	9000	7500
53	7	FL-46	8	13000	10.0	11000	7000
Comparative Example 7	Comparative 1	FL-1	8	3000	10.0	2500	900
8	Comparative 2	FL-1	8	2000	10.0	1500	200
9	Comparative 3	FL-1	8	4500	10.0	3500	600

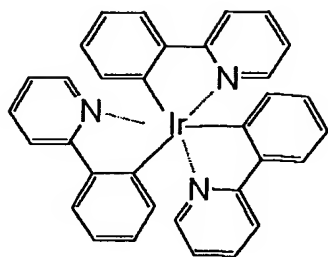
<Example 54>

A device having the structure shown in Fig. 3 was prepared.

On a transparent conductive support substrate
5 similar to that in Example 1, a chloroform solution of a compound represented by the following structural formula was applied into a film of 20 nm in thickness by a spin-coating method, resulting in a hole-transporting layer 5.



Furthermore, the spiro compound represented as the exemplified compound No. 2 and a compound represented by the following structural formula (weight ratio of 100 : 5) were deposited into a film
15 with a thickness of 20 nm by the vacuum evaporation method to form a luminescent layer 3. The film formation was performed under the conditions in which the degree of vacuum at the time of evaporation was 1.0×10^{-4} Pa and the film formation rate was 0.2 to
20 0.3 nm/sec.



Furthermore, an electron-transporting layer 6 was formed by forming a film of 40 nm in thickness from bathophenanthroline (BPhen) by the vacuum
5 evaporation method. The film formation was performed under the conditions in which the degree of vacuum at the time of evaporation was 1.0×10^{-4} Pa and the film formation rate was 0.2 to 0.3 nm/sec.

10 Next, after forming a cathode 4 in the same manner as in Example 1, the resulting product was sealed.

When a direct current voltage of 8 V was applied on the device obtained in this way with an
15 ITO electrode (anode 2) provided as a positive electrode and an Al-Li electrode (cathode 4) provided as a negative electrode, the current was caused to flow into the device at a current density of 10.5 mA/cm² and green-colored luminescence at a luminance
20 of 9000 cd/m² was observed.

Furthermore, when the current density was kept at 7.0 mA/cm² and the voltage was applied for 100 hours, the initial luminance of 7500 cd/m² changed to

a luminance of 6500 cd/m² after 100 hours, indicating small deterioration of luminance.

<Examples 55 to 63>

5 Devices were prepared and evaluated in the same way as that of Example 54, except that the exemplified compound shown in Table 4 was used in place of the exemplified compound No. 2. The results are shown in Table 4.

<Comparative Examples 10 to 12>

10 Devices were prepared and evaluated in the same way as that of Example 54, except that the comparative compounds No. 1 to No. 3 were used in place of the exemplified compound No. 2. The results are shown in Table 4.

Table 4

Example No.	Exemplified compound No.	Initial stage		Current density (mA/cm ²)	Durability	
		Applied voltage (V)	Luminance (cd/m ²)		Initial luminance (cd/m ²)	Luminance after 100-hour (cd/m ²)
Example 54	2	8	9000	7.0	7500	6500
55	4	8	9500	7.0	8000	6500
56	6	8	7000	7.0	6000	5000
57	12	8	8000	7.0	6000	5500
58	14	8	14000	7.0	11000	9000
59	15	8	10000	7.0	9000	7500
60	20	8	10000	7.0	8000	7000
61	22	8	9500	7.0	8000	7500
62	29	8	15000	7.0	13000	10000
63	30	8	8500	7.0	8000	7000
Comparative Example 10	Comparative 1	8	1300	7.0	900	300
11	Comparative 2	8	1000	7.0	900	100
12	Comparative 3	8	2500	7.0	2000	700

<Example 64>

A device having the structure shown in Fig. 1 was prepared.

On a transparent conductive support substrate
5 which was similar to that of Example 1, a solution prepared by dissolving 0.050 g of a spiro compound represented by the exemplified compound No. 1 and 1.00 g of poly-N-vinyl carbazole (a weight average molecular weight = 63,000) in 80 ml of chloroform was
10 applied into a film of 120 nm in thickness by a spin-coating method (rotation speed = 2000 rpm) to form an organic layer (a luminescent layer 3).

Next, after forming a cathode 4 in the same manner as in Example 1, the resulting product was
15 sealed.

When a direct current voltage of 10 V was applied on the device obtained in this way with an ITO electrode (anode 2) provided as a positive electrode and an Al-Li electrode (cathode 4) provided
20 as a negative electrode, the current was caused to flow into the device at a current density of 8.0 mA/cm² and blue-colored luminescence at a luminance of 1900 cd/m² was observed.

Furthermore, when the current density was kept
25 at 5.0 mA/cm² and the voltage was applied for 100 hours in the nitrogen atmosphere, the initial luminance of 1000 cd/m² changed to a luminance of 850

cd/m² after 100 hours, indicating small deterioration of luminance.

<Examples 65 to 68>

5 Devices were prepared and evaluated in the same way as that of Example 64, except that the exemplified compound shown in Table 5 was used in place of the exemplified compound No. 1. The results are shown in Table 5.

<Comparative Examples 13 to 15>

10 Devices were prepared and evaluated in the same way as that of Example 64, except that the comparative compounds No. 1 to No. 3 were used in place of the exemplified compound No. 1. The results are shown in Table 5.

Table 5

Example No.	Exemplified compound No.	Initial stage		Durability		
		Applied voltage (V)	Luminance (cd/m ²)	Current density (mA/cm ²)	Initial luminance (cd/m ²)	Luminance after 100-hour (cd/m ²)
Example 64	1	10	1900	5.0	1000	850
65	7	10	2500	5.0	1600	1400
66	19	10	1400	5.0	1000	800
67	20	10	1800	5.0	1400	1300
68	24	10	1900	5.0	1200	950
Comparative Example 13	Comparative 1	10	300	5.0	200	No luminescence
14	Comparative 2	10	200	5.0	150	No luminescence
15	Comparative 3	10	550	5.0	400	50

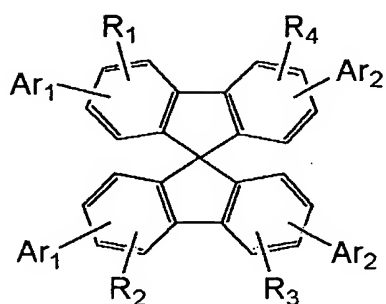
As described with reference to the embodiments and the examples, the organic luminescence device using the spiro compound represented by the general formula [I] or the general formula [II] provides
5 luminescence with a high luminance by the application of a low voltage and is excellent in durability.

In particular, the organic layer containing the spiro compound of the present invention is excellent as an electron-transporting layer and is also
10 excellent as a luminescent layer.

Furthermore, the device can be prepared by using a vacuum evaporation method or a casting method, so that the device having a large area can be easily prepared at a comparatively low cost.

CLAIMS

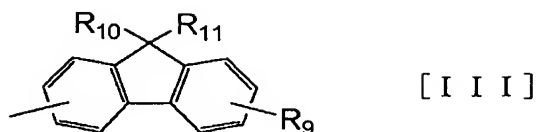
1. A spiro compound represented by the following general formula [I]:



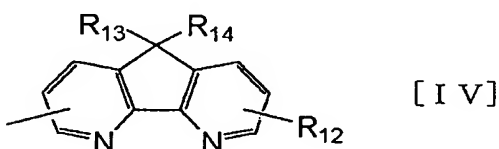
[I]

5 (wherein R_1 , R_2 , R_3 , and R_4 represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a
 10 substituted amino group, a cyano group, or a halogen atom, and R_1 , R_2 , R_3 , and R_4 may be identical or different from each other; and Ar_1 and Ar_2 represent a substituted or unsubstituted condensed polycyclic aromatic group or a substituted or unsubstituted
 15 condensed polycyclic heterocyclic group, which may be identical or different from each other.)

2. A spiro compound according to Claim 1, wherein at least one of Ar_1 and Ar_2 is a condensed
 20 polycyclic aromatic group represented by one of the following general formula [III] to [IX]:

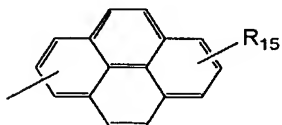


(wherein R₉ represents a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a substituted amino group, a cyano group, or a halogen atom; and R₁₀ and R₁₁ represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, or a substituted or unsubstituted heterocyclic group, which may be identical or different from each other);

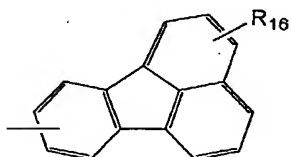


(wherein R₁₂ represents a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a substituted amino group, a cyano group, or a halogen atom; and R₁₃ and R₁₄ represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or

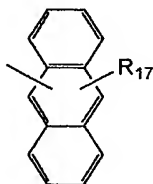
unsubstituted aryl group, or a substituted or unsubstituted heterocyclic group, which may be identical or different from each other); and



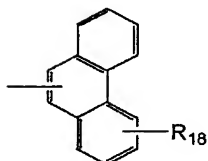
[V]



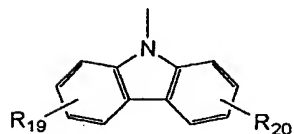
[VI]



[VII]



[VIII]



[IX]

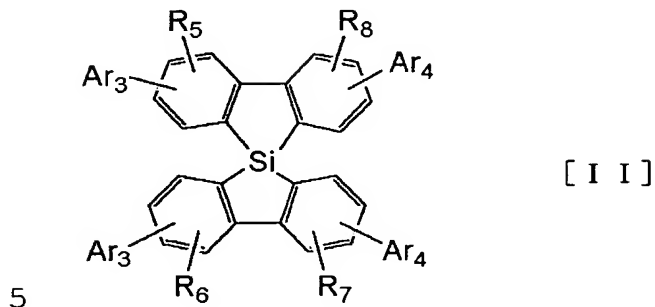
5

(wherein R_{15} to R_{20} represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a substituted amino group, a cyano group, or a halogen

10

atom.)

3. A spiro compound represented by the following general formula [II]:



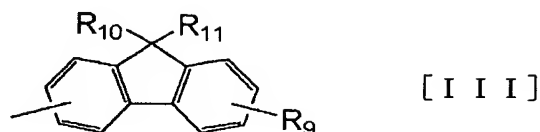
(wherein R₅, R₆, R₇, and R₈ represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a substituted amino group, a cyano group, or a halogen atom, and R₅, R₆, R₇, and R₈ may be identical or different from each other; and Ar₃ and Ar₄ represent a substituted or unsubstituted condensed polycyclic aromatic group or a substituted or unsubstituted condensed polycyclic heterocyclic group, which may be identical or different from each other.)

10

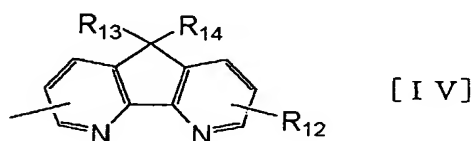
15

4. A spiro compound according to Claim 3, wherein at least one of Ar₃ and Ar₄ is a condensed polycyclic aromatic group represented by one of the following general formula [III] to [IX]:

20

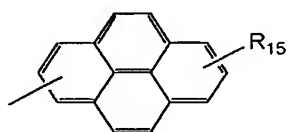


(wherein R₉ represents a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a substituted amino group, a cyano group, or a halogen atom; and R₁₀ and R₁₁ represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, or a substituted or unsubstituted heterocyclic group, which may be identical or different from each other);

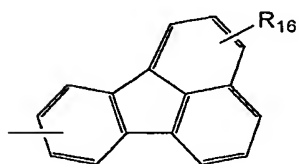


(wherein R₁₂ represents a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a substituted amino group, a cyano group, or a halogen atom; and R₁₃ and R₁₄ represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or

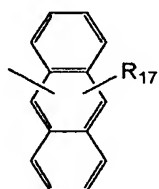
unsubstituted aryl group, or a substituted or unsubstituted heterocyclic group, which may be identical or different from each other); and



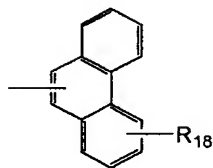
[V]



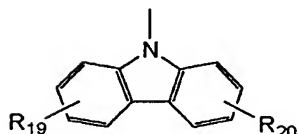
[VI]



[VII]



[VIII]



[IX]

5

(wherein R_{15} to R_{20} represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a substituted amino group, a cyano group, or a halogen

10

atom.)

5. An organic luminescence device comprising at least a pair of electrodes including an anode and a cathode and one or a plurality of layers containing an organic compound sandwiched between the pair of electrodes, wherein at least one of the layers containing the organic compound contains at least one spiro compound according to Claim 1.

10

6. An organic luminescence device comprising at least a pair of electrodes including an anode and a cathode and one or a plurality of layers containing an organic compound sandwiched between the pair of electrodes, wherein at least one of the layers containing the organic compound contains at least one spiro compound according to Claim 3.

15

7. An organic luminescence device according to Claim 5, wherein at least an electron-transporting layer or a luminescent layer among the layers containing the organic compound contains at least one of the spiro compounds.

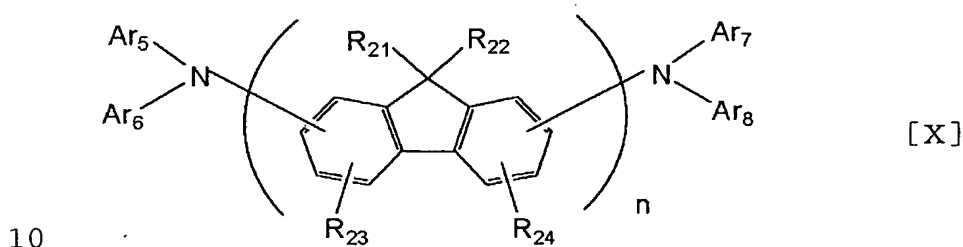
20

8. An organic luminescence device according to Claim 6, wherein at least an electron-transporting layer or a luminescent layer among the layers

25

containing the organic compound contains at least one of the spiro compounds.

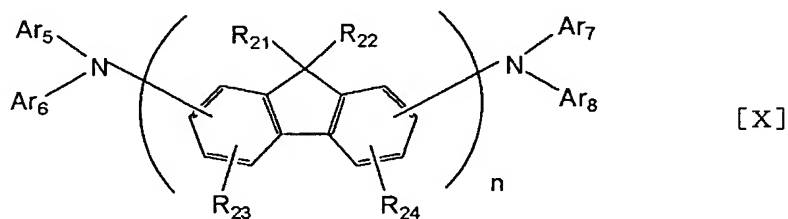
9. An organic luminescence device according to
 5 Claim 5, wherein at least a luminescent layer among the layers containing the organic compound contains at least one of the spiro compounds and a fluorene compound represented by the following general formula [X]:



(wherein R_{21} and R_{22} represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, or a
 15 substituted or unsubstituted heterocyclic group, R_{21} themselves or R_{22} themselves, which are bonded to different fluorene groups, may be identical or different from each other, and R_{21} and R_{22} that are bonded to the same fluorene group may be identical or
 20 different from each other; R_{23} and R_{24} represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a

substituted or unsubstituted heterocyclic group, a cyano group, or a halogen atom, and R_{23} themselves or R_{24} themselves, which are bonded to different fluorene groups, may be identical or different from each other, and R_{23} and R_{24} that are bonded to the same fluorene group may be identical or different from each other; Ar_5 , Ar_6 , Ar_7 , and Ar_8 represent a substituted or unsubstituted aromatic group, a substituted or unsubstituted heterocyclic group, a substituted or unsubstituted condensed polycyclic aromatic group, or a substituted or unsubstituted condensed polycyclic heterocyclic group, which may be identical or different from each other, and Ar_5 and Ar_6 as well as Ar_7 and Ar_8 may be bonded with each other to form rings, respectively; and n represents an integral number of 1 to 10.)

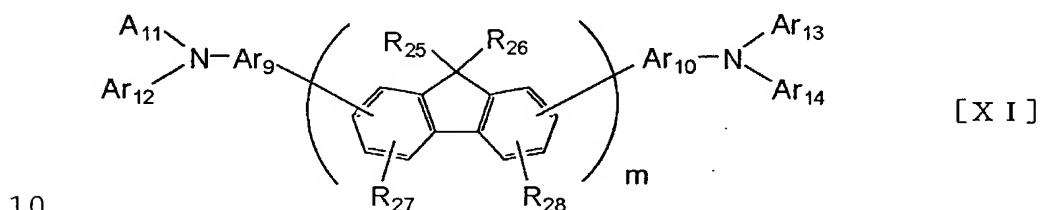
10. An organic luminescence device according to Claim 6, wherein at least a luminescent layer among the layers containing the organic compound contains at least one of the spiro compounds and a fluorene compound represented by the following general formula [X]:



(wherein R_{21} and R_{22} represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, or a substituted or unsubstituted heterocyclic group, R_{21} themselves or R_{22} themselves, which are bonded to different fluorene groups, may be identical or different from each other, and R_{21} and R_{22} that are bonded to the same fluorene group may be identical or different from each other; R_{23} and R_{24} represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a cyano group, or a halogen atom, and R_{23} themselves or R_{24} themselves, which are bonded to different fluorene groups, may be identical or different from each other, and R_{23} and R_{24} that are bonded to the same fluorene group may be identical or different from each other; Ar_5 , Ar_6 , Ar_7 , and Ar_8 represent a substituted or unsubstituted aromatic group, a substituted or unsubstituted heterocyclic group, a substituted or unsubstituted condensed polycyclic aromatic group, or a substituted or unsubstituted condensed polycyclic heterocyclic group, which may be identical or different from each other, and Ar_5 and Ar_6 as well as Ar_7 and Ar_8 may be bonded with each other to form

rings, respectively; and n represents an integral number of 1 to 10.)

11. An organic luminescence device according to
 5 Claim 5, wherein at least a luminescent layer among the layers containing the organic compound contains at least one of the spiro compounds and a fluorene compound represented by the following general formula [XI]:



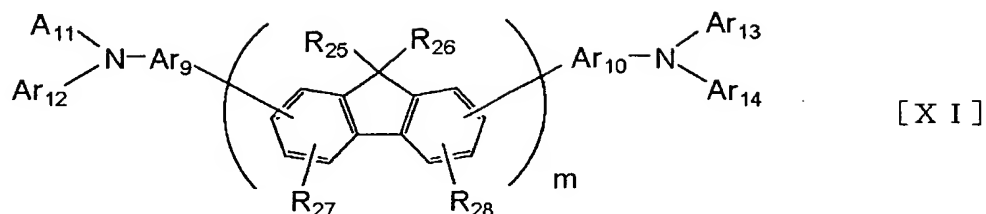
(wherein R_{25} and R_{26} represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, or a substituted or unsubstituted heterocyclic group, R_{25} themselves or R_{26} themselves, which are bonded to different fluorene groups, may be identical or different from each other, and R_{25} and R_{26} that are bonded to the same fluorene group may be identical or
 15 different from each other; R_{27} and R_{28} represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a
 20

cyano group, or a halogen atom, and R₂₇ themselves or R₂₈ themselves, which are bonded to different fluorene groups, may be identical or different from each other, and R₂₇ and R₂₈ that are bonded to the same fluorene group may be identical or different from each other; Ar₉ and Ar₁₀ represent a substituted or unsubstituted divalent aromatic group or a substituted or unsubstituted divalent heterocyclic group, which may be identical or different from each other; Ar₁₁, Ar₁₂, Ar₁₃, and Ar₁₄ represent a substituted or unsubstituted aromatic group, a substituted or unsubstituted heterocyclic group, a substituted or unsubstituted condensed polycyclic aromatic group, or a substituted or unsubstituted condensed polycyclic heterocyclic group, which may be identical or different from each other, and Ar₁₁ and Ar₁₂ as well as Ar₁₃ and Ar₁₄ may be bonded with each other to form rings, respectively; and m represents an integral number of 1 to 10.)

20

12. An organic luminescence device according to Claim 6, wherein at least a luminescent layer among the layers containing the organic compound contains at least one of the spiro compounds and a fluorene compound represented by the following general formula [XI]:

25



(wherein R_{25} and R_{26} represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, or a substituted or unsubstituted heterocyclic group, R_{25} themselves or R_{26} themselves, which are bonded to different fluorene groups, may be identical or different from each other, and R_{25} and R_{26} that are bonded to the same fluorene group may be identical or different from each other; R_{27} and R_{28} represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a cyano group, or a halogen atom, and R_{27} themselves or R_{28} themselves, which are bonded to different fluorene groups, may be identical or different from each other, and R_{27} and R_{28} that are bonded to the same fluorene group may be identical or different from each other; Ar_9 and Ar_{10} represent a substituted or unsubstituted divalent aromatic group or a substituted or unsubstituted divalent heterocyclic group, which may be identical or different from each other; Ar_{11} , Ar_{12} ,

Ar₁₃, and Ar₁₄ represent a substituted or unsubstituted aromatic group, a substituted or unsubstituted heterocyclic group, a substituted or unsubstituted condensed polycyclic aromatic group, or
5 a substituted or unsubstituted condensed polycyclic heterocyclic group, which may be identical or different from each other, and Ar₁₁ and Ar₁₂ as well as Ar₁₃ and Ar₁₄ may be bonded with each other to form rings, respectively; and m represents an integral
10 number of 1 to 10.)

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FIG. 1

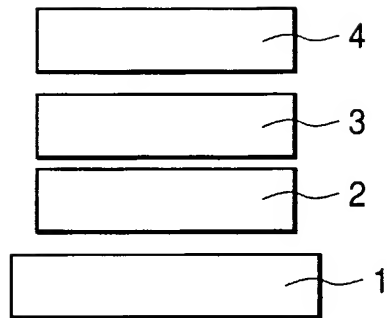
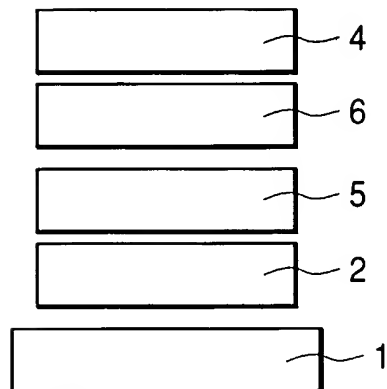


FIG. 2



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FIG. 3

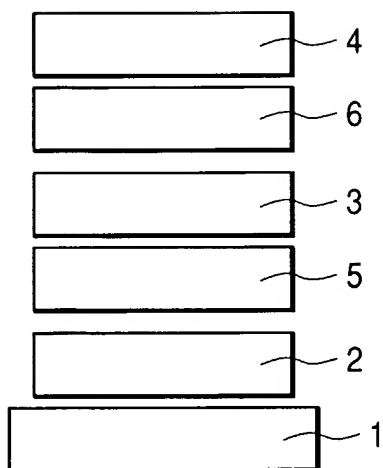
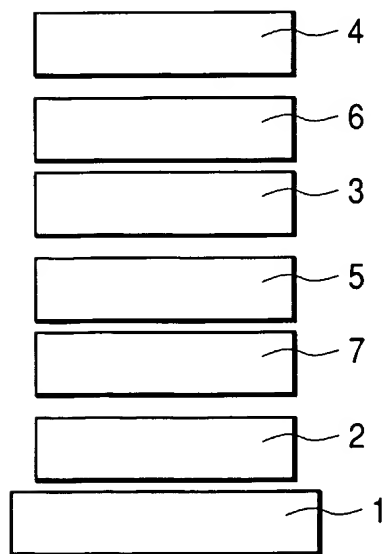


FIG. 4



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FIG. 5

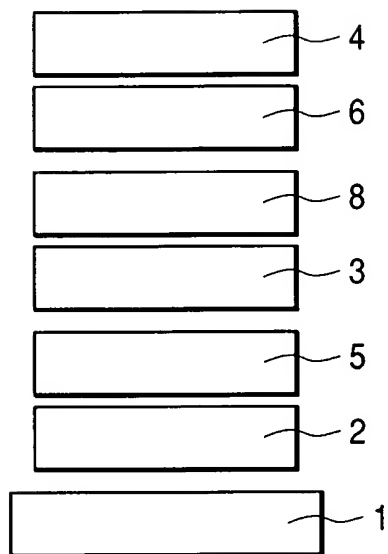
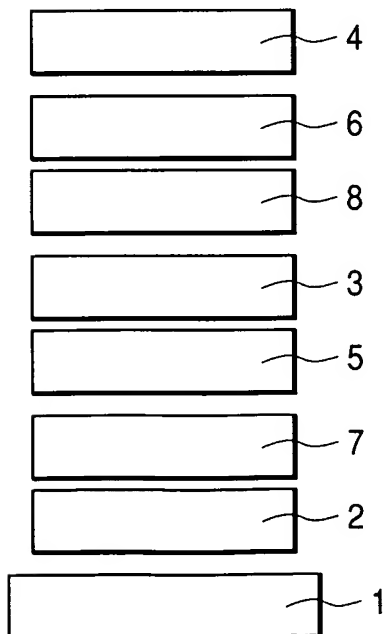


FIG. 6



INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP03/10258**A. CLASSIFICATION OF SUBJECT MATTER**Int.Cl⁷ **C07C13/72, 25/22, 255/52, C07D209/86, 219/02, 471/04, C07F7/08, 7/12, C09K11/06, H05B33/14, 33/22**

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

Int.Cl⁷ **C07C13/72, 25/22, 255/52, C07D209/86, 219/02, 471/04, C07F7/08, 7/12, C09K11/06, H05B33/14, 33/22**Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
Japanese Utility Model Gazette 1926-1996, Japanese Publication of Unexamined Utility Model Applications 1971-2001, Japanese Registered Utility Model Gazette 1994-2001, Japanese Gazette Containing the Utility Model 1996-2001

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

CAPLUS (STN), CAOLD (STN), REGISTRY (STN)**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X A	JP 2002-75645 A (KABUSHIKI KAISHA HANDOUTAI ENERGY KENKYUSYO) 2002.03.15, CLAIM 1 (FAMILY:NONE)	1, 2, 5, 7 9, 11
X A	WO 02/43449 A1 (TORAY INDUSTRIES, INC.) 2002.05.30, PAGE 35, EXAMPLE 5 & JP 2002-222697 A & EP 1341403 A1	1, 5, 7 2, 9, 11
X A	WO 96/17035 A1 (HOECHST AKTIENGESELLSCHAFT) 1996.06.06, PAGE 33, BEISPIEL 16 & DE 4442050 A1 & EP 793699 A1 & CN 1170425 A & JP 10-509996 A & ES 2125056 A1 & US 6329082 A	3, 6, 8 4, 10, 12

☐ Further documents are listed in the continuation of Box C.☐ See patent family annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search

05.09.03

Date of mailing of the international search report

30.09.03

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**4H 9357**